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Indoor Air Quality: Causes, Controls and Consequences

By

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In collaboration with
National Air Quality Testing Services Ltd

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Declaration

This thesis has not been submitted in support of an application for another degree at this or any other university. It is the result of my own work and any work done in collaboration is specifically indicated. Many of the ideas in this thesis were the product of discussion with my supervisory team; Prof Duncan Whyatt, Dr Andrew Sweetman and Douglas Booker.

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Abstract

This thesis aimed to understand the causes, controls and consequences of managing indoor air quality with an emphasis on ventilation throughout, and considerations to energy efficiency. This was achieved through three complimentary sets of experiments.

First this thesis analysed particle number concentrations associated with a series of discrete cooking events to evaluate the efficacy of different types of ventilation in "real world" domestic settings. We identified and characterised 128 discrete cooking events and observed large increases (up to 10^6 particles/cm^3) in particle number concentration in response to these events. A series of key metrics were adopted to enable comparisons to be made between different source and ventilation combinations which revealed that natural ventilation was the most effective means of reducing particle number concentrations in terms of time to background.

Second, we replicated these discrete cooking experiments in a specialist test facility to quantify the energy penalties associated with attempts to improve indoor air quality through use of ventilation. We found that energy penalties are modest (0.082–0.193 kWh) if a period of window opening was restricted to no more than 20 minutes, and that the indoor air quality benefits from this are significant in terms of particle removal. We found that the energy penalties associated with mechanical extract ventilation were even lower for such a period (0.063kWh), and that mechanical ventilation provides the best means of meeting the dual objectives of good indoor air quality and energy efficiency.

Third, we investigated the prevalence of volatile organic compounds within buildings across a university campus to assess the association between volatile organic compounds concentrations and sustainable building standards. We concluded that there were no associations between sustainable building standard ratings and volatile organic compound concentrations, which could result from a lack of indoor air quality related incentives. We suggest a framework for future sustainable building assessment that not only considers ventilation for improving building sustainability and indoor air quality, but also combines continuous total volatile organic compound measurements with detailed speciation.

This thesis was supported by NAQTS who provided access to portable, state-of-the-art V2000 air quality monitoring units. We reflect on the value of such instrumentation and the role it may play in raising public awareness of indoor air quality issues in public and private settings.

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List of Abbreviations and Acronyms

AER: Air Exchange Rate

AUC: Area Under Curve

BREEAM: Building Research Establishment Environmental Assessment Method

BTEX: Benzene, Toluene, Ethylbenzene and Xylene

CO: Carbon Monoxide

CO_2: Carbon Dioxide

EH: Energy House

GC-MS: Gas Chromatography Mass Spectrometry

IAQ: Indoor Air Quality

IOELVs: Indicative Occupational Health Exposure Limits

PM: Particulate Matter

NDIR: Non-Dispersive Infrared

NO_2: Nitrogen Dioxide

PKC: Peak Concentration

TD: Thermal Desorption

RTE: Rate of Decay

TTB: Time to Background

TTP: Time to Peak

TVOC: Total Volatile Organic Compounds

UFP: Ultrafine Particles

VOC: Volatile Organic Compound

WELs: Workplace Exposure Limits

1.0. Introduction

1.1. Background Context

1.1.1. What is Indoor Air Quality?

"Indoor Air Quality" (IAQ) has no universal or standard definition. In general, IAQ is related to pollutants (e.g., biological, chemical and physical) within indoor environments that can affect the health of occupants, but definitions can vary depending on perspectives of the human user, the characteristics of indoor space and the sources contributing to the indoor air pollution (Steinemann et al., 2017). Brown (2019) defines IAQ as "what we experience as the temperature, humidity, ventilation and chemical or biological contaminants of the air inside non-industrial buildings" whereas the US Environmental Protection Agency (EPA) define IAQ as: "the air quality within and around buildings and structures, especially as it relates to the health and comfort of building occupants" (Steinemann et al., 2017).

Historically, studies of IAQ have been largely overshadowed by studies of outdoor or ambient air quality. However, in the western world, we spend the majority of our time indoors (> 90% of our time) where we are exposed to various pollutants (Isaxon et al., 2015). There is growing public awareness about the risks associated with poor IAQ particularly in homes and workplaces (Bernstein et al., 2008). We know indoor air pollution may cause or aggravate illnesses (Daisey et al., 2003, Mendell, 2007), increase mortality (WHO, 2010), and have major economic and social impacts (Fisk and Rosenfeld, 1997, Fisk et al., 2011). Indoor air is a dominant exposure route for humans and IAQ plays a major role with regard to public health (Sundell, 2004). Logue et al. (2011) estimated the effect in disability-adjusted life years per person per year

(µDALY/p/year) from all sources attributable to IAQ excluding second-hand smoke and radon and found it to be in the range between the health effects of road traffic accidents (4000 µDALY/p/year) and heart disease (11,000 µDALY/p/year) (Guyot et al., 2018). Of late, there has been a growing interest in IAQ, and this is reflected in the increasing number of studies in this area. The over-arching topic of this thesis is IAQ, and this introductory chapter will summarise some of the current knowledge in this area which will be later discussed in the literature (Chapter 2).

1.1.2. Sources and Characterisation

IAQ is considered a subset of indoor environmental quality (IEQ) that also includes factors such as lighting, ergonomics, acoustics, and temperature in addition to pollutants (Steinemann et al., 2017). Many different factors contribute to the overall quality of air in an indoor environment. Indoor pollution sources that release gases or particles into the air are the primary cause of IAQ problems. Published research has clearly identified the major sources and types of pollution (gaseous and particulate pollutants) in the indoor environment. We know that indoor environments represent a mixture of indoor and outdoor pollutants, with outdoor pollutants typically associated with vehicular traffic and industrial activities, entering indoor environments by infiltration and/or through natural and mechanical ventilation systems (Cincinelli and Martinelli, 2017). Indoor pollutants originate inside the building, from building materials and furnishings, activities undertaken within the building (including the use of combustion appliances, heating systems, and the storage and application of cleaning and consumer products) and the behaviour and presence of occupants (microbial and metabolic emissions) (Seppänen, 2008; Han et al., 2010; Kumar et al., 2016; Cincinelli and Martinelli, 2017; Salvador et al., 2019). IAQ can also vary according to i) the building characteristics e.g.,

type of foundation, presence or absence of mechanical ventilation, airtightness, and envelope integrity and ii) the characteristics of the outdoor environment (Lavesseur et al., 2017).

In many countries there is no law or body that regulates IAQ even though people typically spend more than 90% of their time indoors and 70% of that time inside their home, with pollutant levels typically several times to several hundred times higher indoors than outdoors (Steinemann et al., 2017; Kruza and Carslaw, 2019; Brown, 2019). That being said the degree of outdoor pollution is strongly dependent on many factors including the country in question. However, this situation is beginning to change as more countries are adopting legislation and standards that target particular pollutants or aspects of IAQ such as ventilation. In the UK for example, a statement issued by the UK Government in 2019 presents a series of IAQ guidelines derived from scientific literature for selected pollutants, to control their levels in the indoor environment through informing discussions on source control and raising awareness, reflecting some progress towards regulation (Public Health England, 2019). In light of the current coronavirus pandemic where we now spend more time indoors, IAQ developments are accelerated and there is increasing pressure on employers to address the issue of providing good indoor air quality.

Exploring indoor sources of air pollutants involves evaluating the processes and products which are used indoors (Brown, 2019). Most air quality studies undertaken by governments and scientists, particularly those of a regulatory nature, use static, expensive, regulation-grade monitoring equipment to measure and assess pollutant concentrations (Lewis et al., 2016). However, equivalent measurements can now be made using low-cost sensors which can be used to measure multiple pollutants

simultaneously at multiple locations (Piedrahita et al., 2014). There has been increasing awareness of the use and practicalities of such low-cost sensors in air quality research, and many studies have evaluated their potential for improving public awareness about IAQ risks and burdens (Lewis et al., 2016). IAQ is, however, difficult to measure and assess due to (i) the lack of consistent metrics, standards and consensus on what constitutes favourable IAQ; (ii) the diversity and complexity of pollutants found indoors that can affect human health and well-being and diversity of issues associated with the range of different environments; (iii) the inadequate understanding of links between pollutant levels indoors, exposure to those pollutants and their effects; (iv) the range of health effects related to indoor pollutant exposures and (v) the lack of requirements to measure and monitor IAQ (Steinemann et al., 2017).

1.2. Problem Statement

Numerous strategies have been presented in the scientific literature that target improvements in IAQ, particularly at the design stage of a building, which is when most IAQ problems arise (Liddament, 1996). Strategies to improve IAQ include source control, ventilation and air cleaning. Source control helps reduce or eliminate individual sources of contamination or emission (Levasseur et al., 2017) and is considered the most effective strategy for improving IAQ (Matson and Sherman, 2004). Adequate ventilation is also required to introduce and circulate fresh air throughout a building and remove or dilute contaminated indoor air to provide a healthy and comfortable living environment (Dimitroulopoulou, 2012). Ventilation rate, expressed as air changes per hour (ACH), is an important determinant for the ingress of ambient air pollutants and removal of indoor pollutants (Breen et al., 2014). Natural ventilation occurs when air infiltrates through unintentional leaks in the building envelope, through intentional

openings (such as open windows, ventilation ducts) and via coupled spaces such as crawlspaces, basements and attics (Liu et al., 2018a). Mechanical ventilation and other measures such as extractors can also help deliver good IAQ (Levasseur et al., 2017). Mechanical ventilation, creating airflow in and out of a building (Seppänen, 2008) adds to the energy demands of a building but can overcome drawbacks of natural ventilation by providing a controlled rate of air change in response to the varying occupant needs and pollutant loads (Liddament, 1996). Air cleaning is another way to improve IAQ such as using high-efficiency particulate air (HEPA) filters.

We know that buildings consume a significant fraction of total energy consumption (a $1/3$ worldwide); thus, are responsible for much of the anthropogenic carbon dioxide emitted that contributes to climate change (Thomsen et al., 2016). Increasing the airtightness of a building saves energy but negatively impacts upon IAQ due to a reduction in the infiltration rate and increase in concentrations of contaminants with indoor sources (Seppänen, 2008; Persily and Emmerich, 2012; Langer et al., 2015; Hamilton et al., 2017; Awbi, 2017). It is not desirable to increase infiltration to improve IAQ as it is positively correlated with heating energy demand (O'Leary et al., 2019b; Dimitrouloupou., 2012). Many organisations are struggling to deal with reducing energy use whilst maintaining acceptable IAQ (Spengler and Chen, 2000; Seppänen, 2008). Several methods have been proposed that target improvements in IAQ without negatively affecting energy consumption such as demand controlled ventilation. Green buildings, certified by various programs (such as BREEAM) typically emphasise efficient use of energy and resources and to a lesser extent, health and indoor air quality. IAQ has been included as one of the default elements of this and other schemes presently in use, which is assessed through awarded credits in the rating systems of such programs. However, concerns have been expressed since IAQ credits contribute to such

a small percentage of credits overall, as to whether the IAQ credits in such schemes are sufficient and allow adequate incentive to pursue these credits (Steinemann et al., 2017).

1.3. Aims and Objectives

The research undertaken in this thesis makes a significant contribution to the broader indoor air science area focusing on and remedying current gaps in understanding and knowledge. The overall aim of the thesis is to evaluate the sources of indoor air pollution and the controls on IAQ within residential and educational micro-environments and to explore the dichotomy between good IAQ and energy efficiency, identifying how we can harmoniously achieve these -sometimes- conflicting objectives. Cooking sources are targeted in this study due to the daily nature of these activities and recent research highlighting the potential harmful nature of pollutants generated through these activities. This aim will be achieved by addressing the following objectives;

- Using low cost, portable monitoring units to measure pollution levels under different ventilation regimes in response to discrete cooking events in;
    a) a specialist test facility and
    b) a selection of households in NW England
- Quantify VOCs between and within sustainably accredited (BREEAM) buildings to assess potential VOC sources and relationships between IAQ and sustainability accreditation.

1.4. Thesis Organisation and Structure

The research in the thesis is concerned with various aspects of IAQ which is of critical importance due to aforementioned negative health effects of poor IAQ. An extensive literature review was conducted prior to undertaking the work in this thesis to evaluate

current research in the field of indoor air science and evaluate the research gaps that could be addressed with future work (Chapter 2). The following chapter (Chapter 3) refers to the instruments used to evaluate IAQ in the research undertaken throughout this thesis. The main chapters (4–6) that comprise the thesis discuss an array of data that was collected between February 2017 and February 2020, with each focussing on one of the objectives outlined in section 1.3.

Chapter 2: This presents a review of the available literature in indoor air science. Topics covered in the literature review include indoor sources and characterisation, controls on pollutant concentrations, measurements and instrumentation, improving IAQ and IAQ-energy efficiency conflicts.

Chapter 3: This chapter presents an overview of the instruments used for indoor air quality monitoring in the work undertaken throughout this thesis and a technical specification of these instruments.

Chapter 4: This chapter presents results from a study that deployed high time resolution air quality instruments with multiple pollutant monitoring capabilities in a selection of households in the NW of England. This study aimed to examine the temporal and spatial particle response to typical episodic household cooking activities and the influences that control cooking emissions, including exposure mitigation to generated particles via natural and mechanical ventilation and housing layout.

Chapter 5: This chapter presents a novel pilot study that replicates cooking activities within the Salford Energy House under different ventilation regimes to assess trade-offs between IAQ and energy efficiency. This pilot study brings new knowledge and understanding to the conflicting objectives of good IAQ and energy efficiency in existing dwellings, through examining temporal variations in indoor pollutant

concentrations from discrete cooking activities. The Energy House provided a unique opportunity to study particle numbers and indoor temperatures at unprecedented temporal resolution. The energy consequences of ventilation are important to consider as these are significant for reducing residential energy use.

Chapter 6: This chapter discusses the results of a study that quantified indoor VOCs within sustainability accredited buildings (BREEAM certified) at a UK university during periods of non-occupancy (building dependent) and occupancy (activity dependent) using a sieve mapping approach. This study aimed to assess the prevalence and concentration of VOCs in high performance buildings with sustainability credentials. This was in order to evaluate sources of VOCs and the relevance of IAQ credits in BREEAM accreditation in order to determine whether the current approach for accreditation is fit for purpose.

Chapter 7: This chapter summarises the main findings of the thesis and based on this offers recommendations for future research in the area of indoor air science.

1.5. Declaration

This PhD was undertaken with a start-up SME, National Air Quality Testing Services (NAQTS, https://www.naqts.com/). Part of the thesis involved testing and utilising their portable V1000/V2000 air quality monitoring units which can simultaneously monitor a variety of gaseous and particulate pollutants and environmental conditions. Chapter 3 presents a technical review of the NAQTS monitoring units. This is followed up in the Appendix with a series of short-term case studies designed to examine its effectiveness for a variety of pollutants.

2.0. Literature Review

2.1. Introduction

Indoor air quality (IAQ) is concerned with concentrations of pollutants and thermal conditions that may negatively affect the health, comfort and performance of a building's occupants (Kubba, 2017). Kubba (2017) states that the four basic factors that influence IAQ are (i) a building's occupants, (ii) a building's HVAC system, (iii) possible pollutant pathways and (iv) possible sources of contaminants. Interest in IAQ began through associations with health. It is well understood that indoor pollutants, even at low concentrations, act as respiratory irritants, toxicants, adjuvants or carriers of allergen (Bernstein et al., 2008) and have led to lung cancer, chronic obstructive pulmonary disease and cardiovascular disease (Spinazze et al., 2019).

Today, the average person spends 90% of their time indoors and 70% of that inside their home (Notman and Carslaw, 2018). Therefore, it is important to understand IAQ and its related problems. This forms the basis for remediating these problems and improving IAQ for the health and well-being of occupants. This literature review discusses current understanding in the overall topic of indoor air science, in order to identify research gaps which, form the basis of the research presented in this thesis.

2.2. Indoor Sources and Characteristics

We know that indoor environments include pollutants from external sources (such as vehicular traffic), which enter by infiltration and/or ventilation systems, and internal sources (Cincinelli and Martinelli, 2017). Indoor contaminants originate from building materials and furnishings, activities undertaken within the building (including the use of combustion appliances, heating systems, and the storage and application of cleaning

solvents and consumer products) and the presence and behaviour of occupants (microbial and metabolic emissions) (Seppänen, 2008; Han et al., 2010; Kumar et al., 2016; Cincinelli and Martinelli, 2017; Salvador et al., 2019). Though invasive, and both time and cost prohibitive (O'Leary et al., 2019a) many researchers have characterised the temporal and spatial patterns of common pollutants (particulate matter, inorganic gases and microbial and chemical volatile organic compounds (Bernstein et al., 2008)) in various indoor microenvironments, which vary between pollutant species and among and within buildings (Sundell et al., 2011). A large number of studies have focussed on domestic residences, particularly kitchens and living rooms due to the amount of time that occupants spend in these microenvironments, and the presence of major indoor sources. Studies of schools and universities, offices and commercial buildings are also relatively common (Vu et al., 2017).

The concentration of individual pollutants indoors depends on (i) emission rates from various indoor sources (ii) rates of transport from outdoors to indoors and (iii) the rates at which they are deposited on indoor surfaces, consumed by indoor chemistry and removed by ventilation/filtration (Weschler and Carslaw, 2018).

2.2.1 Particles

Particle number concentrations (PNC) vary from low values ($<10^3$ particles/cm^3) in clean indoor environments to high values ($>10^4$ particles/cm^3) during active periods of occupancy and very high values ($>10^6$ particles/cm^3) in the presence of intense indoor sources (Bo et al., 2017; Isaxon et al., 2015).

Recent studies highlight the main indoor particle sources relate to cooking activities including frying, sautéing, toasting and baking (Long et al., 2000). However, smoking has also been associated with high PNC, as has the use of household appliances

including gas-fired ranges and ovens, kerosene heaters, wood-stoves and fireplaces, along with other more general activities including incense burning, walking and vacuuming (Long et al., 2000). Combustion sources tend to elevate ultrafine (UFP, <0.1 µm), fine or accumulation mode (AMP) (0.1–2.5 µm), and nanoparticle concentrations. In contrast, activities resulting in resuspension (e.g., physical movement) tend to elevate coarse particle concentrations (2.5–10 µm) (Long et al., 2000; Howard-Reed et al., 2003; Bo et al., 2017). There is an extensive body of literature analysing particulate pollution, particularly from cooking-related activities. Episodic sources such as cooking are the cause of peak concentrations and variability in exposure among buildings (Bhanger et al., 2011).

Residential environments are perhaps the most commonly studied environments. Most airborne particles in residences, when expressed as PNC, are generated by residents themselves through combustion/thermal related activities (Isaxon et al., 2015; Fantke et al., 2017); cooking, wood-burning, candles and smoking. Highest UFP concentrations have been associated with burning pure wax candles [241,000 particles/cm^3], cooking (frying meat, electric and gas stove), smoking, and the use of electric heaters (Pederson et al., 2001; Afshari et al., 2005; Gehin et al., 2008; Bhanger et al., 2011). In contrast vacuuming, sweeping, use of non-terpene cleaning products and ironing without steam on a cotton sheet [550 particles/cm^3] have not appeared to lead to a notable enhancement of PNC (Bhanger et al., 2011). Though not considered in this review, the use and application of chemical cleaners can generate UFP and coarse particles, which is particularly attributed to the oxidation and condensation of VOCs contained with them (Vu et al., 2017).

Cooking is an important part of daily food preparation in residential and commercial settings for the safety and enhancement of a substantial number of food products, to reduce food-borne illnesses, and to alter the composition of food products (Hager and Morawicki, 2013). It is also one of the most significant indoor sources of particles and organic gas emissions (Dennekamp et al., 2001; Wallace et al., 2004; Wheeler et al., 2011; Kearney et al., 2011; Wallace and Ott, 2011; Rim et al., 2012; Klein et al., 2019) which can reach hazardous concentrations in the kitchen space and elsewhere in the internal environment. Therefore, it can contribute significantly to personal exposure and adversely affect health if concentrations are not maintained below health-based thresholds (Logue and Singer, 2014; Lunden et al., 2015; O'Leary et al., 2019a). High cooking temperatures and cooking practices more generally generate large amounts of smoke which may cool and nucleate to form UFPs that dominate number concentration but contribute negligibly to particle mass concentration (Lai and Ho, 2008; Nazaroff, 2018). It has long been recognised that cooking can create high concentrations of visible aerosol indoors but cooking it is now also being considered a significant component of particles outdoors (Abdullahi et al., 2013). Cooking emission studies have been carried out in real world environments where emissions are influenced by numerous factors (e.g., room arrangement, building materials, outdoor infiltration, other combustion devices, ventilation and cooking methods) (Abdullahi et al., 2013). Emission studies have also been conducted in controlled environments where measurements are influenced by few factors, mainly the fuel and food used (Abdullahi et al., 2013). Fewer studies focus on cooking in commercial settings, though some of this work has been undertaken (Lee et al., 2001; Ots et al., 2016; Gysel et al., 2018).

Numerous researchers have investigated cooking emissions and influencing factors but reported emission rates are highly variable due to the many influencing factors and

complications associated with real-world environments. Examples include toasted bread (9.5 ± 10.8 mg/min), fried chicken breast (15.2 mg/min) and deep-fried French fries (0.34 ± 0.03 mg/min) (O'Leary et al., 2019a). It has been said that peak concentrations may be more important for health effects than long-term concentration averages (Garrett et al., 1998). Peak PNCs from cooking have been found to be higher than reported outdoor peak concentrations by at least an order of magnitude. Zhang et al. (2010) reported increases in UFP exposure up to 550 times that of background levels during cooking. Concentrations of between 90,000–150,000 particles/cm^3, 400,000 particles/cm^3 and 200,000–300,000 particles/cm^3 have been found from scrambling eggs (Li et al., 1993), from deep frying tortillas on a gas stove burner followed by baking in the oven and from sautéing shrimp on a gas stovetop burner (Wallace and Ott, 2011) respectively. Dennekamp et al. (2001) and Afshari et al. (2005) noted that UFPs rose to a peak of 150,000 particles/cm^3 simply by turning on 4 gas rings (or in the latter frying meat on an electric stove) and then to a peak of 590,000 particles/cm^3 when frying bacon.

The cooking of individual components, rather than full meals, may not be representative of typical home meal preparation. O'Leary et al. (2019a) studied emission rates and source strengths for complete meals. Likewise, He et al. (2004) characterised cooking emissions, finding variable emission rates of 0.03–2.78 mg/min and peak PNC of between 16,000 and 180,000 particles/cm^3. Increased number and volume concentrations during dinnertime may, in part, have been due to the increased time of cooking (He et al., 2004). More research is required to assess PNC emissions from cooking full meals.

The Home Observations of Microbial and Environmental Chemistry (HOMEChem) study has made some progress in this area in investigating the influence of everyday

activities on the emission, chemical transformation and removal of gases and particles indoors through extensive collaborative research (Farmer et al., 2019). Sequential experiments repeating similar activities throughout the day were interspersed with periods of enhanced ventilation (window opening) to investigate emissions (Farmer et al., 2019). Layered experiments replicated cooking and cleaning activities throughout the day with no interspersed window opening to simulate real-life use of a home (Farmer et al., 2019). During cooking events, large particle enhancements occur, which by number are largely in the ultrafine mode, with a substantial fraction owing to chemical species related to cooking oils (Farmer et al., 2019) (Figure 2.1). Substantial mass changes are also observed in the accumulation and super-micron modes (Farmer et al., 2019).

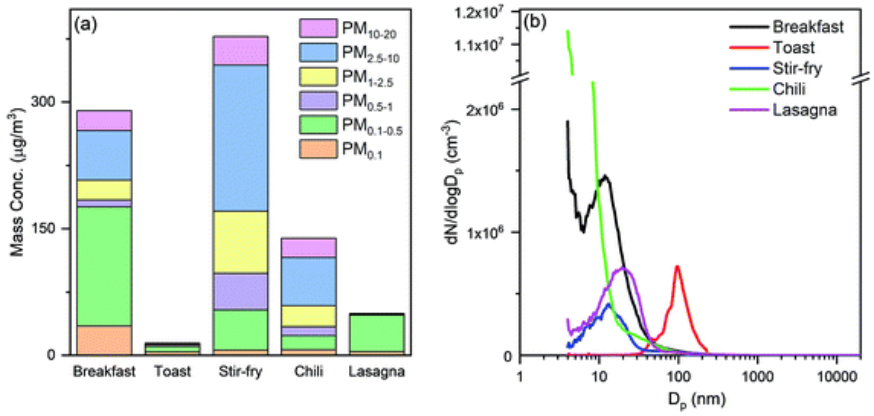

Figure 2.1: Particulate response to typical cooking activities in the HOMEChem Study. (**a**) particulate matter mass concentration measurements from typical cooking activities during the HOMEChem experiments according to particle size and (**b**) measured particle concentration plotted against particle size for individual cooking events (Farmer et al., 2019).

Most UFPs are said to be produced in response to the flame or heating elements, rather than the pots, pans or food (Wallace et al., 2008). Nonetheless more recent work highlights there is evidence that the cooking equipment itself can influence emission rates, especially when there is absorbed organic matter on the surface of pans (O'Leary et al., 2019a). Indeed, particle emission rates from and during the processes used in cooking (e.g., frying, roasting, grilling, boiling) are seen to span several orders of

magnitude; affected by ingredients, procedures, cooking style or setting and cooking temperature as well as air exchange rates and oxidant precursor levels (Abdullahi et al., 2013; O'Leary et al., 2019a; Klein et al., 2019). Dry, water-based, and oil-based cooking processes have very different emission rates, with oil-based methods, such as frying and grilling, having the highest rates (up to 30 and 90 times the ambient concentration) (He et al., 2004). Higher particle numbers and mass concentrations have also been found at higher cooking temperatures by some researchers. Siegmann and Sattler (1996) show that the PNCs increased twofold with an increase of the oil temperature from 223 to 256 °C. Evidence suggests that cooking ingredients influence $PM_{2.5}$ emissions, and oil type (smoke point, composition, and water content) is perhaps the most significant (O'Leary et al., 2019a). The effects of non-essential additives, e.g., seasonings, on emission rates have also been investigated. Further contradicting the previous assertion food type has been found to be important, with the fat content of foods and their emission rate being highly correlated (O'Leary et al., 2019a). Fuel type is also significant. Higher emission rates are reported when using gas burners rather than electric hobs by Buonanno et al. (2009) but not by others.

Particle size distributions have been extensively studied. It is well known that combustion- generated particles are considerably smaller than 2.5 μm, often smaller than 1 μm, justifying the use of number concentration of ultrafine particles as a more relevant metric than mass when determining residential exposure to combustion related particles (Isaxon et al., 2015). Most of the particles emitted from gas/electric stoves have been seen to be less than 0.04 μm with the peak PNCs occurring around 0.005 μm to 0.006 μm (Wallace et al., 2008; Rim et al., 2012) and a slight shift in size during frying of bacon (0.05–0.1 μm) (Dennekamp et al., 2001). Wallace et al. (2006) highlighted a shift towards larger particle sizes for more complex dinnertime cooking,

such as using gas burners, stir-frying and pan-frying (0.036–0.04 µm) and using a gas oven (broiling fish, baking potatoes) (0.045–0.046 µm). Singer and Delp (2018) summarised cooking sources that produced large numbers of particles at size fractions <0.3 µm; heating water on a gas stove, cooking a pizza in a gas oven, cooking pancakes over medium heat, and toasting bread in a well-used electric toaster oven, and those that generated large quantities of $PM_{2.5}$; heating oil in a wok on gas or electric burners, frying bacon, toasting 4 slices of bread in a toaster oven, and stir-frying green beans on a gas burner.

Particles in indoor air are influenced by various physical and chemical processes which change their physical characteristics, chemical composition and concentrations (Bekö et al., 2020). Particle exposure from indoor sources is a function of the source strength and losses due to air exchange, filtration, coagulation, and deposition (Wallace et al., 2019). All previous studies assessing particle count data from cooking show similar temporal trends. Concentrations from the onset of cooking are initially low, then rise steeply with the rate of increase depending on many factors including the cooking method, the relative location between the source and the sampling area and the indoor airflow (buoyancy and convection) (Lai and Ho, 2008). Concentrations increase over time as the cooking continues indicating ongoing emissions of particles. High peaks in concentration are quickly generated (Afshari et al., 2005), with maximum concentrations reached between a few minutes and a half hour (Afshari et al., 2005; Klein et al., 2019).

After cooking stops, concentrations decrease towards background levels at a rate that is usually exponential (with the rate of decrease of particle concentration with time proportional to the concentration), determined by air change rate (governed by

ventilation, mainly that provided by a kitchen fan and/or natural ventilation) and deposition on interior surfaces (and where number concentration is higher than 10,000–20,000 particles/cm^3, by coagulation) (Isaxon et al., 2015). Typically, the increase of the particle concentration immediately after the onset of cooking is more rapid than the observed decay once cooking has ceased. During the decay, the total number concentration decreases with time and the particle size distribution moves toward larger particle sizes as the aerosol ages (Wallace, 2000; Abt et al., 2000; Dennekamp et al., 2001).

The lifetime of the cooking aerosol particles in the kitchen has been reported to vary between 4–6 h (Hussein et al., 2006). PNCs in adjacent living spaces can also be affected. Wan et al. (2011) noted that during cooking average number concentrations of UFPs and AMPs were 20–40 times and 10 times greater than background levels in the kitchen and living room respectively. PNCs then remained elevated after cooking for up to 90 and 60 minutes in the kitchen and living room. The average number mean diameter of UFPs and AMPs in the living room was about 10 nm larger than that in the kitchen during cooking, highlighting coagulation effects (Wan et al., 2011).

Laboratory and field-based studies show combustion-related particles contain a host of organic and inorganic material (Morawska and Zhang, 2002; Klein et al., 2019) including alkanes, fatty acids, alkanones, sterols, polycyclic aromatic hydrocarbons and heterocyclic amines (Abdullahi et al., 2013) and more complex oxidised organic molecules such as sorbic and lactic acid (Farmer et al., 2019). Experimental work has characterised cooking emissions and found that whilst frying processes are the main driver of larger and unsaturated aldehyde emissions, terpenes are mostly emitted due to condiment use (Klein et al., 2019). Farmer et al. (2019) found that organic aerosol dominated the

submicron mass during cooking, and, while variable between meals and throughout cooking, was dominated by components of hydrocarbon character and low oxygen content, similar to cooking oil. Emitted particles evolve throughout cooking, becoming more oxygenated and nitrogenated when food is added to cooking oil (Farmer et al., 2019).

2.2.2. Organic Compounds

Volatile organic compounds (VOCs thereafter), carbon-based chemicals which contain a range of chemical species (including saturated and unsaturated hydrocarbons, carbonyls, alcohols, ethers, esters, furanoids, amines, siloxanes and sulphides) with a high vapour pressure at room temperature (above 0.01 kPa at 20 °C) (Goodman et al., 2017), are prevalent indoor air pollutants. The most well-documented VOCs are benzene, toluene, ethylbenzene, and xylene (BTEX) compounds and 1,3,5-Trimethylbenzene (1,3,5-TMB). Documented indoor sources include consumer products and building materials; wood (MDFs and particle boards), thermal and acoustic insulations, carpets, paints, coatings, industrial solvents, adhesives, fireproof materials, PVC, flooring, and furnishings (Shaw et al., 2005; Cacho et al., 2013). Advances in construction and changes in building materials, including the use of recycled material and more synthetic materials (Jones, 1999) have introduced more organic gases indoors (Spengler and Chen, 2000). Even green consumer products and building materials can emit potentially hazardous VOCs (Goodman et al., 2018).

Formaldehyde, whose indoor concentrations typically exceed outdoor concentrations, is often treated separately as it is not detected by gas chromatographic methods that quantify VOCs (Shaw et al., 2005). Sources include additive degradation in wood-based building materials, furniture, sealants, combustion, and chemical reactions (Destaillats et al., 2006; Singer et al., 2006; Kruza and Carslaw, 2019). Recent research has shown

that semi-volatile organic compounds (SVOCs) can also be emitted from building materials; flooring, furniture, electronics, plastic items, textiles, cleaning, and cosmetic products (Kristensen et al., 2019).

Elevated VOC concentrations associated with difference source types and different activities; building materials, furnishings, and household products inside the living space; occupants and their episodic activities; chemical processes; and transport from outdoors or connected spaces have distinctive characteristics and are a concern for residential IAQ (Farmer et al., 2019). High-baseline concentrations indicate continuous indoor emissions from building materials and furnishings (Kristensen et al., 2019) When indoor sources are absent, concentrations are typically lower than outdoors, as VOCs are expected to adsorb on surfaces or be chemically destroyed (Yurdakul et al., 2017).

In the past, concern has focused on primary emissions from building materials and furnishings (Liu et al., 2019) which may decay in days or weeks, but secondary emissions due to ageing of the material persist over longer periods (Sundell, 2004; Prasaukas et al., 2016). Compared to older buildings, recently constructed buildings have shown increased carbonyl concentrations and total VOCs likely due to increased ventilation in older dwellings and lower emissions from older building materials (Molloy et al., 2012; Langer et al., 2015). Reasonably good IAQ in newer buildings is generally attributed to higher air exchange rates owing to mechanical ventilation. Continuous emission patterns for many compounds indicate ongoing chemical processes such as decomposition and oxidation (Liu et al., 2019). Liu et al. (2019) suggest slow decomposition of the wooden building envelope is a major source for acetic acid, formic acid, and methanol, which accounted for 75% of the total continuous

indoor emissions. When compared to conventional buildings, the IAQ of energy efficient buildings has been marked by high concentrations of terpenes and hexaldehyde, likely attributed to wood or wood-based products (Derbez et al., 2017). Langer et al. (2015) reported significant sources of total volatile organic compounds (TVOCs) in passive houses and formaldehyde in conventional houses.

Observed airflow patterns highlight that air pollutants can enter occupied spaces from coupled zones (e.g., crawlspaces, attic) (Liu et al., 2019). Liu et al. (2019) found substantial upward inter-zonal airflows, with most VOCs observed in the living spaces of residences being emitted from sources directly into the living space and negligible transport from outdoor and coupled spaces. Sleeping environments are usually characterised by lower ventilation rates (Bekö et al., 2010) which tends to promote pollution accumulation (Canha et al., 2017). Mattresses, pillows and bed linens are often treated with flame-retardants and contain residual detergent components and other substances such as SVOCs that can be re-suspended during sleep and impact human health (Canha et al., 2017; Boor et al., 2017).

Intermittent emissions from occupants and their activities produce short-term enhancements in VOC concentrations (Liu et al., 2019). Human occupants in buildings enhance pollution owing to emissions of alcohols, hydrocarbons, aldehydes and ketones, with concentrations in the range of ppb to ppm including acetone, acetate and pentanal, from skin oils and shedding of skin flakes, rich in skin oil, and breath (Verielle et al., 2016; Weschler and Carslaw, 2018; Kruza and Carslaw, 2019; Farmer et al., 2019). Their concentration in the indoor environment depends on the volume of the indoor space, the air change rate, the number of individuals indoors and individual variations such as diet (Kruza and Carslaw, 2019). Experimental studies show the sources, behaviour,

and time series of VOCs, including oxidized organic acids, following human occupancy and occupant activities (cooking and cleaning) are complex (Farmer et al., 2019).

Many studies note that VOC exposures are affected by an individual′s activity. In the RIOPA study, Su et al. (2013) found most VOC exposures (66–78%) in non-smoking households occurred indoors. VOCs with the highest average concentrations in Michigan residences included aromatics (benzene, toluene and xylenes), which are solvent-related and in household products, paints, adhesives, synthetic fragrances, evaporated fuel and vehicle emissions, alkanes (n-C7–13 and methyl cyclohexane) and terpenes (d-limonene and α-pinene), constituents of cleaning products, air fresheners and fragrances (Chin et al., 2014). Bari et al. (2015) similarly attributed VOCs in residences to household products (44%), combustion and environmental tobacco smoke (10.5%), deodorizers (8.4%) and off-gassing of building materials (5.9%). Presence of a carpet, use of a dishwasher, washing clothes, painting or varnishing floors and furniture within the last 12 months caused elevated concentrations of VOCs in 60% of homes studied by Bari et al. (2015). Residential air exchange rate (AER, or ventilation) has been negatively associated with indoor levels of toluene, xylenes, styrene, chloroform and monoterpenes (Su et al., 2013). In terms of environmental factors, ambient humidity and wind speed were negatively associated with indoor VOC levels (Su et al., 2013).

Household, consumer, and maintenance products such as air fresheners, cleaning products and personal care products can emit VOCs (such as monoterpenes, acetaldehyde, acetone, toluene, xylenes, decane, undecane, dodecane and ethanol) during usage (Goodman et al., 2018; Massolo et al., 2010; Jenkin et al., 2000; Derbez et al., 2017). In HOMEChem, mopping with pine-scented cleaner raised limonene levels,

while mopping with bleach solution raised chloroform levels (Farmer et al., 2019). Indoor emissions of cyclic volatile methylsiloxane (cVMS) (octamethylcyclotetrasiloxane (D4), decamethylcyclopentasiloxane (D5), and dodecamethylcyclohexasiloxane (D6)) associated with personal care products have also been studied (Nazaroff et al., 2015; Yang et al., 2018).

Educational institutions are commonly studied indoor environments (Akal et al., 2015; Allou et al., 2008; Chan et al., 2007; Godwin and Batterman, 2007; Goodman et al., 2018; Park et al., 2014; Solomon et al., 2008; Yurdakul et al., 2017) with a focus on primary and high schools as they house high density populations of young people who are particularly vulnerable to air pollutants. Human emissions are important in highly occupied spaces, e.g., classrooms, more so now energy efficiency measures are making buildings more airtight (Kruza and Carslaw, 2019). Zhong et al. (2017) examined VOCs in conventional schools and schools built to high sustainability credentials. Most VOC concentrations were low (mean <5 $\mu g/m^3$) and the most prevalent were aromatic compounds e.g., toluene, benzene, m/p-xylene and 1,2,4-trimethylbenzene (Zhong et al., 2017). BTEX, terpene and formaldehyde concentrations were positively correlated with the presence of vinyl and wood floor materials and negatively correlated (along with TVOCs) with carpeted floors, whilst VOCs (except formaldehyde) were associated with the presence of science class materials (Zhong et al., 2017). Building type (conventional vs high performance) did not appear to have a significant influence on VOC concentrations (Zhong et al., 2017).

Several international studies have studied IAQ in university buildings but generally university buildings have drawn less attention (Yurdakul et al., 2017). Chan et al. (2007) studied VOCs across a university campus and attributed the main VOCs; toluene and

benzene to ingress from outdoors. Solomon et al. (2008) found environmental tobacco smoke was a main factor in indoor pollution at the University of Bremen and pollutants associated with cleaning products and materials exhibited higher concentrations indoors than outdoors. Goodman et al. (2018) studied the prevalence and concentration of VOCs at an Australian University across campus services, restrooms, renovated offices, a green building, meeting areas and classrooms, revealing the most prevalent VOCs (ethanol, d-limonene and formaldehyde) had links with building materials, furnishings and fragranced consumer products.

Due to the use, application and storage of volatile solvents and chemicals, relatively higher VOC concentrations have been detected in many (university) buildings housing laboratories (Valavanidis and Vatista, 2006; Park et al., 2014; Yurdakul et al., 2017). Park et al. (2014) found concentrations of 11 VOCs within laboratory buildings were significantly higher (mean: 185 $\mu g/m^3$) than those of non-laboratory buildings (mean: 12.1 $\mu g/m^3$) owing to the presence and use of laboratory chemicals; ethanol, acetone, methylene chloride, n-hexane and chloroform. Even when using fume hoods organic materials can be a source of VOCs when heated (Yurdakul et al., 2017). In addition, fume hoods may exacerbate outdoor air pollution (Park et al., 2014). Rumchev et al. (2003) and Valavanidis and Vatista (2006) investigated IAQ in university laboratories in Australia and Athens respectively (Park et al., 2014) showing occupants can be exposed to higher levels of TVOCs compared to non-laboratory environments, although these can be reduced by air conditioning.

2.2.3. Inorganic Gases

Combustion activities are responsible for elevating levels of some inorganic gaseous pollutants indoors, especially in residences. Carbon monoxide (CO) is formed from

incomplete fuel combustion, and as such has been positively correlated with gas cooking (Molloy et al., 2012). Carbon dioxide (CO_2) is exhaled by humans and, in the absence of combustion activities, is the greatest contributor to indoor concentrations (Jones, 1999). Typical concentrations range from 700–2000 ppm but can exceed 3000 ppm when unvented appliances are used (Jones, 1999). Indoor ozone and ammonia sources include air purifiers and laser printers, human emissions, pets and household products respectively (Sutton et al., 2000; Zhang and Smith, 2003; Bernstein et al., 2008; Salonen et al., 2018). Indoor ammonia concentrations can be higher than outdoor concentrations (1–5 ppb) (Ampollini et al., 2019). Ammonia has been related to (thanksgiving) cooking where concentrations have been found to range between 24 and 130 ppb. A rapid ammonia increase as the oven is opened suggests thermal decomposition of amino acids in meat proteins is responsible (Ampollini et al., 2019). Indoor ozone concentrations are highly variable, with specific indoor sources including air purifiers, laser printers and photocopiers (Salonen et al. 2018). Nitrogen compounds form during combustion. Primary indoor sources of nitrogen oxides are unvented fuel burning appliances, heating appliances and tobacco smoking (Vilcekova, 2011). Nitrogen oxides play key roles in ozone formation.

2.3. Controls on Concentration

2.3.1. Building Characteristics

Occupant behaviour is an important determinant of pollutant concentrations, which varies between and within buildings. Indoor pollutant concentrations are also influenced by building age, size, type, and use, which vary considerably, both in terms of geographic location and method of construction. Other influencing factors include the characteristics of interior materials, the airtightness of the building envelope and type

of ventilation system (Langer and Bekö, 2013; Lavesseur et al., 2017). UK Building regulations Parts L and F are related to indoor air quality and give guidance of air tightness and ventilation respectively (UK Government, 2014a). Proper building envelope design and occupant behaviour can limit infiltration of contaminants from the outside, while regular maintenance and inspection of buildings can prevent the deterioration of building materials that are not designed to be exposed to the elements (Levasseur et al., 2017). Residential buildings constructed in the past two decades tend to be more airtight than older buildings, with lower air exchange rates (Weschler, 2009) preventing ingress of outdoor pollutants. Conversely, newer buildings often have higher concentrations of airborne pollutants that are generated in the indoor environment from materials and activities (Fortenberry et al., 2019). Building operators and designers are encouraged to avoid low ventilation rates unless alternative effective measures are employed (Sundell et al., 2011). When designing buildings, it is important to account for local pollution-generating processes by locating them in separate rooms (Seppänen, 2008).

Levasseur et al. (2017) stated that we must keep designing and building 'performant' buildings (e.g., green and net-zero energy buildings) that promote good IAQ, and energy efficiency. Since the 1970s significant momentum toward energy conservation in buildings has led to energy related building codes and has resulted in the tightening of building envelopes reducing air infiltration (Mudarri, 2010). Regulations targeted towards energy efficiency are also included in the UK Building Regulations Approved Documents (UK Government, 2014a). Newer designs, construction practices and building materials for "green" buildings and the use of "environmentally friendly" products can potentially reduce chemical exposure, but this is not always the case (Zhong et al., 2017).

2.3.2. Indoor Chemistry

Chemical reactions occurring in materials, on the surface of materials or in the gas phase have a great influence on the chemical composition of indoor air (Uhde and Salthammer, 2007; Weschler and Carslaw, 2018). These chemical processes and their relation to those occurring outdoors must be well-understood (Bekö et al., 2020). From 1991–2010 more than 250 peer-reviewed publications addressed reactions among indoor pollutants (Weschler, 2011) including oxidation, hydrolysis, acid/base, photolysis and decomposition (Weschler and Carslaw, 2018). Most studies, at least initially, were undertaken in controlled chambers, often neglecting the influence of occupancy. Some researchers have focused on particles being oxidised by ozone during episodic activities such as cooking, and surface-mediated ozonation driven by clothing and skin (e.g., Fortenberry et al., 2019). Another trending topic is chemistry in hidden building spaces and how this influences chemistry in occupied spaces (Weschler and Carslaw, 2018). The field of indoor air chemistry is moving forward rapidly, accelerated to some extent by the Alfred P Sloan Foundation's "Chemistry of Indoor Environment" Program (Bekö et al., 2020). In addition, INDAIRPOLLNET (2018–2022) is also addressing the current state of knowledge of indoor air pollution, with an emphasis on indoor air chemistry (Indairpollnet, 2019). With increasing use, more studies into reactions of green materials containing nanoparticles are also envisaged (Weschler and Carslaw, 2018).

The reaction between ozone (O_3) and the terpenes (a class of VOCs) has been extensively explored in the literature, given the common occurrence of ozone in outdoor air and terpenes in indoor environments, which react fast enough to compete with air change rates (Long et al., 2000; Wainman et al., 2000; Weschler, 2004; Fan et al., 2003; Fan et al., 2005; Destaillats et al., 2006). Fan et al. (2003) found that when O_3 was added

to terpenes or a mixture 23 VOCs including terpenes (α-pinene and d-limonene), reaction products included aldehydes, organic acids and submicron particles. The mechanism for the reaction between O_3 and the terpenes was identified as O_3 addition to a >C=C< bond of the terpene to form a primary ozonide, which reacts further to form hydroxyl carbonyls for example (Fan et al., 2003). Fiedler et al. (2005) shows formaldehyde is a product of this reaction increasing from 13 $\mu g/m^3$ (no ozone) to 40 $\mu g/m^3$ in the presence of 40 ppb ozone. Other experimental studies support these findings, with reaction products including unsaturated VOCs; the hydroxyl (OH), hydroperoxy (HO_2), organic peroxy and nitrate (NO_3) radicals, and Criegee intermediates (Weschler and Carslaw, 2018). Fan et al (2005) and (Chen and Hopke, 2009) similarly discuss how adding O_3 to a mixture of VOCs led to the formation of submicron particles.

Earlier studies did not investigate reactions between ozone and skin surface lipids because they took place in unoccupied areas (Weschler, 2016) but now such studies are prevalent. Human occupants contribute to reactive chemicals. Breath is a significant source of reactive chemicals indoors; containing isoprene, nitric oxide (NO) and ammonia (Weschler and Carslaw, 2018). Skin oils are transferred onto surfaces that humans contact, and skin flakes can deposit on horizontal surfaces (Weschler and Carslaw, 2018). Squalene has been considered the most important skin surface lipid that readily reacts with ozone (Weschler, 2016) entering buildings from outdoors via ventilation and infiltration (Kruza and Carslaw, 2018). Through the presence of occupants indoors and decreases in oxidant levels, the formation of nitrated organic species, potentially toxic compounds, can be affected (Kruza and Carslaw, 2018).

The time available for chemical reactions indoors is determined by the building ventilation rate, which influences reactant, ozone, and seed particle concentrations, and by dry deposition of the reactants (Langer et al., 2008; Waring and Siegel, 2010; Weschler and Carslaw, 2018). For gas phase reactions to influence indoor environments, the time scale of the reaction must be competitive with air change (Weschler and Carslaw, 2018). Emerging research focuses on low- or zero-energy buildings, which often have low ventilation rates providing more time for gas-phase chemistry (Weschler and Carslaw, 2018). Salvador et al. (2019) quantified the influence of ventilation on occupant-related indoor air chemistry. Exposure to noxious products of ozone/human chemistry can be reduced by decreasing ventilation during periods with high outdoor ozone levels. Turning off the ventilation overnight or on weekends may lead to the accumulation of certain pollutants with indoor sources but could limit the extent that ozone-derived products are formed (Salvador et al., 2019). Time constraints do not apply to surface reactions, unless they involve airborne particles (Weschler and Carslaw, 2018). Higher rates of terpene emission (Sarwar et al., 2003) and higher rates of ventilation have been shown to increase O_3/terpene reaction rates and reaction products (Coleman et al., 2008) but the latter will also dilute these products (Kruza and Carslaw, 2018). Higher outdoor particle concentrations have been seen to cause higher indoor 'seed' particle concentrations, increasing organic aerosol concentrations (Sarwar et al., 2003).

Other reactions are considered including base-catalyzed hydrolysis of plasticizers and personal care products (Weschler and Carslaw, 2018). Reactions with ammonia are also considered; ammonia reacts with acidic gases such as H_2SO_4, HNO_3, and HCl and with bleach, enabling the formation of secondary aerosol mass as ammonium sulfate

$(NH_4)_2SO_4$, ammonium nitrate $(NH_4)NO_3$, ammonium chloride NH_4Cl, and chloramines (NCl_3, $NHCl_2$) (Farmer et al., 2019).

2.3.3. Outdoor: Indoor Exchange

Historically, outdoor air pollution has been the focus of air quality research because of public awareness and acknowledgements of associated health impacts. It is important to consider the impact that outdoor (ambient) air quality has on IAQ and several researchers have evaluated this. The impact, of course, depends on where the building is located (e.g., city or countryside) and how airtight it is. In the absence of indoor pollution sources, studies show a general trend of higher outdoor than indoor concentrations (Bo et al., 2017). Generally higher ventilation rates cause indoor air to become more like local outdoor air.

The main outdoor sources include emissions from vehicles, coal and gas-fired power stations, industry, agriculture, domestic heating systems and atmospheric reactions (Bo et al., 2017). Common sources of prevalent VOCs, benzene and toluene include petroleum and vehicle exhaust and their presence in the indoor environment indicates close proximity to heavily trafficked roads (Chan et al., 2007). VOCs in the atmosphere can react with UV-rays contributing to tropospheric photochemical ozone formation over wide areas (Park et al., 2014). Whereas indoor activities intermittently influence indoor PNCs, outdoor particle concentrations continuously influence indoor (and indoor baseline) concentrations (Bhanger et al., 2011). Coarse particles are generally associated with natural sources whilst fine, ultrafine and nano-scale particulates are generally associated with anthropogenic sources (Bo et al., 2017).

2.4. Indoor Air Quality Testing, Monitoring and Modelling

2.4.1. Questionnaires

Qualitative assessments of IAQ and the presence, duration, frequency and patterns of exposures involves addressing occupant satisfaction in buildings through the distribution of questionnaires/surveys (Wargocki et al., 2000a; Fang et al., 2004; Nieuwenhuijsen, 2004; Lai et al., 2004; Clausen and Wyon, 2008). These are not considered further within this review because we largely focus on quantitative analysis in our investigations in this thesis.

2.4.2. Quantifying Pollutant Concentrations

A wide range of techniques have been used to evaluate IAQ across a range of micro-environments. Most air quality studies, particularly those of a regulatory nature undertaken by governments and scientists, use static monitoring stations equipped with certified reference instruments (Lewis et al., 2016). These analysers are typically large, heavy and expensive, costing between £5000 and £60,000 (Mead et al., 2013). These instruments are subject to strict maintenance and calibration routines to ensure high quality data and comparability between sites (Castell et al., 2017). They also require infrastructure such as secure and temperature-controlled enclosures (Piedrahita et al., 2014).

In the case of VOCs, their total concentration (TVOC) can be measured or individual species can be quantified, which is more desirable because of the effects of some individual components (Ras et al., 2009). European standards ISO 16000-5:2015 (Sampling Strategy for Volatile Organic Compounds) and ISO 16000-6:2011 (Determination of VOCs in Indoor Air) are particularly relevant to the analysis of VOCs in indoor air. Sampler devices can quantify cumulative VOC levels but cannot track temporal patterns (Castell et al., 2017). To quantify individual species of VOCs sample

concentration, followed by separation by gas chromatography and detection by sensitive GC detectors, is required (flame ionisation detection (FID), electron capture detection (ECD) or mass spectrometry (MS)) (Helmig and Vierling, 1995; Ras et al., 2009). Proton-transfer reaction mass spectrometry (PTR-MS), which works based on reactions of H_3O^+ ions, also allows individual VOCs to be monitored with high sensitivity (Wang et al., 2015). Whilst GC-FID, GC-MS and PTR-MS are highly sensitive and linear in response, these instruments are very expensive and not portable and thus less suitable for field analysis but have been used in outdoor field campaigns (Wang et al., 2015).

For carbonyl compounds, air is sampled onto 2,4-dinitrophenylhydrazine (DNPH)-treated silica cartridges from stable derivatives in situ. Sampling time varies in previous studies from 7 hours (at 1200 mL/min) (Goodman et al., 2018) to 168 hours (Geiss et al., 2009). High performance liquid chromatography with ultra-violet detection (HPLC-UV) is the most common analytical technique. Validated methods are based on ISO standards including 16000-3 Indoor Air: Part 3 Determination of formaldehyde and other carbonyl compounds in indoor air and test chamber air – Active sampling method for the determination of carbonyl compounds (International Organization for Standardization, 2020).

Liu et al. (2019) analyse a full spectrum of VOCs by PTR-ToF-MS through continuous monitoring campaigns. The OFFICAIR and AIRMEX studies measured VOCs with a passive sampler. In the former study, VOCs were analysed by TD coupled with capillary GC-MS (Mandin et al., 2017). This technique was also used in the RIOPA study (Zhong et al., 2017) and numerous others (Roberts, 2012; Vette et al., 2013; Csobod et al., 2014; Sakai et al., 2017). In the AIRMEX study GC-FID was used for analysis. Similarly, Derbez et al. (2014) measured VOCs by a passive sampler, and provided quantification

through GC, MS and FID. Goodman et al. (2018) analysed VOCs in university buildings using an automated thermal desorber (ATD) and a Hewlett Packard GC/MS/FID in accordance with US EPA method TO–17. All of these studies performed carbonyl analysis using HPLC-UV detection (Derbez et al., 2014) in accordance with US EPA Method TO–11A (Goodman et al., 2018).

Approaches used for measuring particulate matter (PM) concentration (gravimetric, microbalance and optical) and size distribution (scanning mobility particle sizer, electrical low-pressure impactor and others) are discussed in the literature. Filter-based gravimetric samplers have been widely used in ambient particle monitoring (Amaral et al., 2015). Microbalance methods, including the tapered element oscillation microbalance (TEOM) analyser are sometimes used in indoor-outdoor studies, but most measurements of time-resolved UFP and $PM_{2.5}$ in indoor environments have been made with photometers, optical particle counters (OPCs) and condensation particle counters (CPCs) based on the principle of light scattering (Amaral et al., 2015; Singer and Delp, 2018). Among size distribution methods, microscopy can provide much information (Amaral et al., 2015). A recent development is the Electrical Low Pressure Impactor (ELPI), which classifies particles according to their aerodynamic diameter (Amaral et al., 2015). Other complete systems of spectrometers for measuring particle mobility diameter include the scanning mobility particle sizer (SMPS) which comprises a Differential Mobility Analyser (DMA) and a Condensation Particle Counter (CPC) and is based on the principle of the mobility of a charged particle in an electric field (Amaral et al., 2015). Aerodynamic particle sizers (APS) measure particle size distributions from 0.5 to 20 μm by determining the time of flight of individual particles in an accelerating flow field (Peters and Leith, 2003).

These methods have been used in numerous studies of IAQ. O'Leary et al (2019a), O'Leary et al. (2019b) and Isaxon et al. (2015) used OPCs in their measurement campaigns to measure $PM_{2.5}$ and particle size distribution in residential environments. Other researchers report the use of a CPC (Fan et al., 2005; Afshari et al., 2005; Kearney et al., 2011; Bhanger et al., 2011) to measure fine and ultrafine particles and SMPS to monitor PNC and particle size distribution (Wallace et al., 2004; Wallace et al., 2019; Hager and Morawska, 2013).

Ventilation metrics include ventilation rate ($m^3 \cdot h^{-1}$), ventilation rate per person ($L \cdot s^{-1} \cdot person^{-1}$) and outdoor air change rate (A, h^{-1}). These can be determined using (i) air flow measurements; (ii) pulse or constant injections of tracer gases; (iii) occupant-generated carbon dioxide (CO_2) or (iv) through a comparison of indoor and outdoor concentrations (Batterman et al., 2017). Tracer gas measurements, based on the mass balance of a tracer gas in a building, have been used to calculate air change rates and airflow characteristics in many US and European homes (Yamamoto et al., 2010; Dimitroulopoulou, 2012; Breen et al., 2014; Liu et al., 2018a). Occupant-generated CO_2 has been widely used since CO_2 is inert, emission sources (people) are present in all buildings and usually well dispersed throughout occupied spaces, and inexpensive and accurate measurement and logging instruments are available (Batterman et al., 2017). The pressurised blower method can also determine AER which determines building envelope leakage (Breen et al., 2014). Pressurisation measurements have been used to calculate inputs for some AER models.

2.4.2.1. Sensors

Low-cost sensors which make autonomous measurements of multiple pollutant parameters at a cost of 100–10,000 USD per observing location (Lewis et al., 2016;

Singer and Delp, 2018) have the potential to take equivalent measurements to reference instruments while capturing additional spatial variability (Piedrahita et al., 2014). Start-up companies have begun producing low-cost air quality monitors housing low-cost sensors which aim to provide information in real-time at a resolution not previously observed (Kumar et al., 2016; Lewis and Edwards, 2016). A number of recent studies have examined the value of low-cost sensors in indoor air science. These studies are mostly assessment based and often evaluate inter-sensor comparability and their performance against reference-grade instrumentation. However, the findings from these studies have been contradictory. The cost and flexibility of deployment of low-cost sensors are often cited as major advantages, however, it is not yet known how useful they will be in the future of indoor air pollution monitoring. Further studies could evaluate the use of these sensors in the field, which is currently missing, and the use of multi-parameter field calibrations to improve reported measurements.

Some of most advanced air quality sensors are seen to recreate general patterns of pollutant behaviour captured by reference instruments over short timescales (Lewis et al., 2016; Smith et al., 2017). Singer and Delp (2018) found four consumer AQ monitors (AirBeam, AirVisual, Foobot, and Purple Air II) provided quantitative or nearly quantitative measurements that were time correlated and within a factor of 2 for most sources investigated and were therefore seen to be of sufficient accuracy and reliability to detect large sources. Two consumer monitors (Air Quality Egg and Awair) responded to most sources but reported mass concentrations less than half of the estimated true values (Singer and Delp, 2018). All the consumer and research monitors in this study substantially under-reported or missed events when much of the emitted mass consisted of particles smaller than 0.3 μm diameter; however, as many UFP sources also emit particles above this size fraction, the monitors could still help reduce UFP exposures

(Singer and Delp, 2018). The performance of these monitors needs to be evaluated in occupied homes and quantified over longer periods.

It is difficult to make trace gas measurements to a usable degree of accuracy and precision and with stability over time (Lewis et al., 2016). Sensor performance is affected by effects such as drifts of zero and calibration slope as well as cross sensitivities and interferences to and with other gases (co-pollutants) and environmental or meteorological parameters (e.g., water vapour, temperature) (Williams et al., 2013; Lewis and Edwards., 2016; Smith et al., 2017; Batterman et al., 2017). Over time the ranking of individual sensors changes since sensors each respond to different environmental conditions with slightly different sensitivities to each parameter (Smith et al., 2017). Lewis et al. (2016) found interference from stable longer-lived gases that were not the target analyte such as CO_2 and H_2 were small, but a high ratio of these co-pollutants to the measurand could cause large artefact responses. Poor agreement between NO_2 electrochemical sensor measurements to reference NO_2 suggests this sensor is also responding to another pollution metric, in this instance ambient CO_2 (Lewis et al., 2016). Mead et al. (2013) shows that although electrochemical sensors used to measure NO and NO_2 agree well with reference techniques (provided cross sensitivities are accounted for) there is interference for O_3 (100%) (Mead et al., 2013). Interferences can be reduced by using filters and co-locating with reference analysers. Castell et al. (2017) used an NO_2 sensor with filter to reduce or eliminate O_3 cross-interference and found no cross-sensitivity with O_3.

Responses induced on each sensor by individual interferences do not change substantially over timescales of seconds to a few hours however they vary considerably over the >6 hour to 1–2-day timescale (Smith et al., 2017). Smith et al. (2017) highlight

that the inter-sensor spread of observed values increased as the sensor signals drifted apart over the 3-week timescale. Time-averaging sensor signals can address short-term random noise but not medium-term drift in sensor sensitivities to measurand or interferences (Smith et al., 2017). Emerging literature highlights the importance of regular (once a day) multi-parameter calibration for individual sensors to be comparable to one another and to reference instrumentation due to their non-linear relationship with cross-interferences (co-pollutants and environmental parameters) and drift over time (Smith et al., 2017). Calibration must be of the target compound and all other possible interferences (Lewis and Edwards, 2016; Smith et al., 2017). Castell et al. (2017) show multivariate field calibration is necessary to reduce bias and measurement errors which is difficult since sensors have sensitivity to surrounding environmental conditions and do not normally have access to in-service reference materials for calibration (Smith et al., 2017). The practicalities of such calibrations conflict with the concept of low-cost sensors (Smith et al., 2017). The inclusion of multiple different sensors in a clustered approach could bring the performance of sensor technologies closer to reference instruments, thereby improving the quality of observations (Smith et al., 2017). Using the median concentration of the cluster of sensor signals largely eliminates variability of individual sensors on the hour-to-day timescale (Smith et al., 2017). The remaining systematic decline in response can be corrected for by linear interpolation between infrequent calibrations (Smith et al., 2017). Emerging literature shows corrections for chemical and environmental factors can be improved using more complex statistical models; partial least squares, neural networks, or Gaussian process emulation (Smith et al., 2017).

Sensor systems for VOCs have a particular attraction due to the expense and practicalities of using GC/MS in the field (Smith et al., 2017) and thus limited

observational datasets of VOCs (Lewis et al., 2016). Sensors could deliver something complementary to existing approaches, a direct measurement with a degraded level of chemical detail with well-resolved time and spatial resolution (Lewis et al., 2016). Lewis et al. (2016) discussed the difficulty in defining what these sensors are responding to when measuring TVOC; the values they are reporting are also not easy to compare to reference instruments (Smith et al., 2017).

2.4.3. Modelling

Indoor air pollutant measurement techniques are unable to measure multiple pollutants at sufficient temporal resolution and with the required specificity in a wide range of buildings to provide a representative understanding of processes occurring indoors (Bekö et al., 2020). Computer simulation techniques have been used to estimate indoor concentrations or exposures and predict the impacts of intervention (O'Leary et al., 2019b). Computational Fluid Dynamics (CFD) modelling techniques can be used to simultaneously predict indoor and outdoor airflows, heat transfer and contaminant distribution and transportation in and around buildings (Zhai, 2006).

2.5. Improving Indoor Air Quality

Studies that evaluate the many ways we can improve IAQ through source control, ventilation and air cleaning are widespread. However, fewer of these consider the practical implications of these solutions in real world situations.

2.5.1. Standards and Guidelines for Indoor Contaminants

Indoor air has not been regulated like outdoor air (Langer and Bekö, 2013). Indoor air standards are not widely reviewed in the literature possibly due to the lack of information about them. A number of countries have described target concentrations for

various indoor pollutants (Harrison, 2002), many of which are adopted or derived from outdoor air contaminant standards set by the WHO and ASHRAE. The UK Air Quality Strategy set targets for reducing ambient concentrations of $PM_{2.5}$ and other pollutants to comply with EU legislation (O'Leary et al., 2019b). In 2019, Public Health England issued IAQ guidelines derived from scientific literature for selected VOCs to control their levels in the indoor environment through informing discussions on source control and raising awareness (Public Health England, 2019). Furthermore, since people are exposed to various substances at work, some of which are potentially harmful, indicative occupational health exposure limit values (IOELVs) have been introduced under the Chemical Agents Directive (98/24/EC) (Health and Safety Executive, 2018) through Workplace Exposure Limits (WELs), which are considered to a limited extent in the literature. Exposure concentrations need to be placed in context with toxicological information and given guidelines accordingly.

2.5.2. Source Reduction and Control

Source control helps eliminate or reduce individual sources of contamination (Levasseur et al., 2017) and is noted as the most effective strategy for improving IAQ (Matson and Sherman, 2004). The history of home heating is a good example, with sealed modern fireplaces being considered more effective at reducing emissions in the living space than older open fireplaces (Guyot et al., 2018). Reducing or eliminating unnecessary pollutants at source and using low pollution products and materials (Seppänen, 2008) is generally considered more effective than diluting pollutant concentrations by ventilation (Dimitroulopoulou, 2012; Guyot et al., 2018). High emission rates can produce poor IAQ irrespective of ventilation characteristics (Nazaroff, 2013). Effective source control can also reduce ventilation energy

requirements (Seppänen, 2008). Optimizing the building envelope (insulation and airtightness) is another method of source reduction, since it limits the occupants' exposure to physical stresses and external contaminants (Lavasseur et al., 2017; Nazaroff, 2018).

Product information on material emissions from manufacturers can be used to predict IAQ in the building design stage through modelling (Altaf et al., 2014). Empirical models, based on analysis of emissions data from environmental chamber or cell testing, have enabled characterisation of VOC emissions from building materials and consumer products (Liu et al., 2013) according to international standards; EN180 16000-9:2006 or EN180 16000-10:2006. Testing of single building products and materials under standard conditions may help reduce VOC emissions, but may not give realistic results due to indoor chemistry (Uhde and Salthammer, 2007). Adhesives and floor coverings could be ranked as low-emitting materials under single product chamber testing but in the real world interactions between these materials could give rise to new chemicals (Uhde and Salthammer, 2007). Empirical models have been seen to be difficult to scale from chamber to building conditions (Xu and Zhang, 2003). Furthermore, emission testing provides little insight into the mechanisms controlling emissions (Liu et al., 2013). Mass transfer theory models can predict VOC emissions for various conditions when physical parameters are known (Xu and Zhang, 2003).

Building materials considered to be better for IAQ include durable materials with clean non-toxic materials, low VOC emissions, low moisture content and moisture absorptivity, and low toxic chemical and fibre content (Spengler and Chen, 2000). Han et al. (2010) found using 'exterior-grade' pressed wood products or coating pressed wood products with polyurethane was better for IAQ than traditional materials.

Reduced concentrations of VOCs in indoor air relative to permissible limits in a study by Vasile et al. (2016) could be explained by the low emissions from relevant internal surfaces finishing or furniture, taking into account that there had been no recent renovations in the monitored spaces. Materials used in older construction were more forgiving of temperature and humidity variations and they often acted as sponges for absorbing contaminants (Spengler and Chen, 2000). Those used in newly constructed buildings have a reduced sink area for contaminant absorption and impervious surfaces are then covered with many non-natural finish products glued in place (Spengler and Chen, 2000). Determining the sorption of building materials is important to quantify IAQ (Yang and Chen, 2001). Equilibrium models assume sorption and desorption are confined on the material surface and an equilibrium is achieved between phases at the interface (Yang and Chen, 2001). Kinetic models take VOC diffusion mechanisms into consideration but are largely based on the assumption that indoor air is well-mixed (Yang and Chen, 2001; Lee et al., 2005).

Labelling and certification of building materials and products concerning their emissions has proven useful in minimising emissions through incentivisation but there is no agreed labelling procedure, only suggestions by relevant associations including Business and Institutional Furniture Manufacturer's Association who attach labels to materials to confirm testing by an independent laboratory and meeting of requirements (Avgelis and Papadopoulos, 2004; Levasseur et al., 2017). Bekö et al. (2020) compares 13 labelling schemes for construction products worldwide. However, whilst no study at present has examined the efficiency of labelling schemes to significantly reduce the occupants' exposure to contaminants, selecting less-emissive materials is still considered an incentive measure to reduce contaminants at the source (Lavasseur et al., 2017). Several countries have adopted legislation regarding aspects of building

construction and IAQ e.g., all construction materials and interior decoration products sold in France must have standardized labels to provide information on VOC emissions (Levasseur et al., 2017).

Occupant behaviour is important. Source control can be achieved through selecting and using low-emitting equipment (for example fuel switching to electric hobs (Wilkinson et al., 2009)) and appliances in pollutant-generating activities. In cooking it is possible to reduce $PM_{2.5}$ emissions by using methods that do not brown or char the food and frying with non-stick pans (O'Leary et al., 2019b). Other methods: replacing oil with liquid margarine and adding salt have a minimal effect on $PM_{2.5}$ emission rates (O'Leary et al., 2019a). Other occupant choices including avoiding smoking indoors, avoiding the use of unvented stoves, fireplaces or space heaters, limiting candle or incense burning indoors, correctly using and storing potentially toxic household and pest control products and avoiding the use of air fresheners, cleaning products and fragrances with a pine or citrus scent (Lavasseur et al., 2017) are important. Fragrance-free policies restricting the use of fragranced products have been implemented in buildings worldwide (Steinemann et al., 2017).

2.5.3. Ventilation and Ventilation Standards

Airflow in houses comprises ventilation through purpose–provided openings, infiltration and exfiltration through adventitious openings, and airflow through mechanical systems (O'Leary et al., 2019b). Adequate airflow or ventilation, involving introducing and circulating fresh air through a building and removing or diluting contaminated indoor air, is needed to provide a healthy and comfortable environment within a building (Dimitroulopoulou, 2012). Ventilation should be sufficient to dilute contaminant concentrations to below harmful thresholds (Spengler and Chen, 2000).

Ventilation rate, expressed as air changes per hour (ACH) or air exchange rate (AER), is an important determinant for the ingress of outdoor air pollutants and removal of indoor pollutants (Breen et al., 2014). A common working hypothesis is that the larger the supplied ventilation rates, the greater the indoor pollutant removal efficiency. This is provided outdoor or supplied air is clean. Increased ventilation may worsen IAQ if there are significant outdoor sources of air pollution or outdoor air pollution burdens. Increasing the ventilation rate is often the first line of defence to improve IAQ (Matson and Sherman, 2004) and has been shown to reduce the proportion of people dissatisfied with poor IAQ (Wargocki et al., 2000b). Designers may specify higher ventilation rates before and during initial occupancy of newly constructed or recently renovated buildings since this period is often accompanied by the presence of strong emission sources (Levin, 1991).

There is no guarantee that an occupant will use installed ventilation so many studies consider infiltration-only as a means of ventilation (O'Leary et al., 2018). Traditionally houses were so "leaky" that air infiltration could provide dilution of indoor-generated pollutants even when windows were closed (Singer et al., 2006) but now infiltration is considered a poor mechanism because infiltration airflow rates are low (due to airtightness) and these rates cannot be increased due to concerns over heating energy demand (O'Leary et al., 2018). While 47% of English houses have a fan in their kitchen, the majority solely rely on infiltration for dilution during the heating season (in the UK, this is usually from October–March) when windows are usually closed (O'Leary et al., 2019b). In these circumstances, occupants are likely exposed to pollutant concentrations that exceed WHO daily indoor and outdoor guidelines (World Health Organisation, 2020). Canha et al. (2017) report that infiltration-only ventilation

(closed doors and windows) has resulted in mean VOC levels above the limit value of 0.6 mg/m^3 established by the legislation.

Natural ventilation (NV) occurs through air infiltration in unintentional leaks in the building envelope, through intentional openings (such as open windows, ventilation ducts) and via coupled spaces such as crawlspaces, basements, and attics (Liu et al., 2018a). NV, driven by wind and thermally-generated pressures has in the past has met ventilation needs (Dimitroulopoulou, 2012). Apart from in the north, the European ventilation system is mainly attributed to uncontrolled air infiltration and natural ventilation (window opening) (Dimitroulopoulou, 2012). NV or window opening, increasingly promoted as an environmentally and economically sustainable practice to meet home cooling requirements, particularly in a warming climate, significantly increases the ventilation rate in dwellings and can prevent under-ventilation even in airtight buildings (Lowe, 2000; Fortenberry et al., 2019). However naturally ventilated buildings are generally seen to be older and constructed from traditional materials which can result in lower pollution loads (Wargocki et al., 2002) and this is important to consider in terms of its efficiency. Human occupancy presents challenges to assessing NV impacts on IAQ (Fortenberry et al., 2019).

Residents play an important role in controlling ventilation rates in their own homes (Dimitroulopoulou, 2012). Efforts from the occupants to manually open windows and control the natural ventilation and their tendency to do so only when perceiving a problem with IAQ or comfort affect the efficiency of natural ventilation (Sundell et al., 2011; Liu et al., 2018a). Natural ventilation consumes little energy and provided the outside air is clean, can provide a larger amount of fresh air than mechanical ventilation (Spengler and Chen, 2000). NV is, however, difficult to control due to reliance on

unreliable driving forces, which can result in periods of insufficient ventilation and periods of over-ventilation and excessive energy waste (heat loss) (Liddament, 1996; Lowe, 2000). Nasir and Colbeck (2013) note that ventilation rates were more stable when the windows were closed than open. Furthermore, whilst window opening can reduce concentrations of some indoor-originating pollutants, it can allow ingress of harmful pollutants from the outdoor environment, including ozone and particulate matter, and increase emission rates of semi- and intermediately volatile species and oxidation products (Canha et al., 2017; Liu et al., 2018a; Kruza and Carslaw, 2018; Fortenberry et al., 2019).

Changes in building design aimed at improving energy efficiency and conversation since the 1970s have led to modern homes and offices becoming more airtight which has reduced exchanges between outdoor and indoor air (Zhang and Smith, 2003). It has been suggested that many modern homes and offices built to tight envelope specifications are under-ventilated and may not provide sufficient outdoor (ventilation) air to dilute indoor-generated contaminants (Mudarri, 2010). Whilst in Britain's temperate climate, houses used to be so leaky that whole-house mechanical ventilation was not economic, as new builds are more airtight, these systems are being installed (Dimitroulopoulou, 2012). Mechanical ventilation and other measures such as extractors can compensate for reductions in NV rate caused by improvements in airtightness (Levasseur et al., 2017). Improving building airtightness without providing additional ventilation leads to lower ventilation rates and poorer IAQ (O'Leary et al., 2019a).

Mechanical ventilation, airflow in and out of a building caused by a fan through intake and/or exhaust vents (Seppänen, 2008), adds to the energy demands of a building but

can provide controlled rates of air change in response to the varying occupant needs and pollutant loads (Liddament, 1996). In colder climates, where houses need to be airtight to conserve heat, whole house mechanical ventilation systems have been installed since NV is not adequate (Dimitrouloupou, 2012). This is also the case in warmer regions where buildings are airtight to reduce energy consumption. Higher ventilation rates have been measured in mechanically ventilated dwellings compared to the naturally ventilated dwellings in many countries (e.g., Netherlands, Portugal, Sweden) (Dimitrouloupou, 2012). Mechanical ventilation systems are becoming installed in more residential buildings, in particular mechanical ventilation with heat recovery (MVHR) and mechanical extract ventilation (MEV) (Sullivan et al., 2012). In the Netherlands, these systems have been fitted to nearly all new homes built in the past 10 years (Sullivan et al., 2012). In Western Europe, the payback time for investments in heat recovery ventilation is significant (Laverge et al., 2011).

Available literature discusses the history of ventilation standards and requirements around the world which receive major attention in building regulations. Building ventilation recommendations were transformed into more rigorous standards in the 20th century (Sundell, et al., 2011). In Europe, the European Committee for Standardization (CEN) is responsible for most standards relating to ventilation including EN13779: Ventilation for Non-Residential Buildings and EN13799: Specific Design Guidelines and Requirements to Ventilation Systems (Olesen, 2011). Minimum ventilation requirements, including passive ventilation plus exhaust provisions for known contaminant sources, are the principle way in which building codes address IAQ concerns (Mudarri, 2010). Hypothetically, the ventilation rate for an indoor space in the absence of any pollutant sources would equal the outdoor air supply rate necessary for human metabolism, which ranges from 0.1 to 0.9 l/s per person (Wargocki et al., 2002).

It is difficult to set ventilation rates that would meet requirements for health in all indoor environments (Wargocki et al., 2002). The rate in both naturally and mechanically ventilated buildings can be affected by time-varying factors including internal heating and cooling loads, outdoor temperature, and indoor-outdoor temperature differences (Godwin and Batterman, 2007). In most European countries the minimal ventilation rate for new buildings with mechanical ventilation is 0.5 air changes per hour (ACH). Sufficiently high ventilation rates are needed so as to not compromise IEQ and cause health, comfort, absenteeism and productivity problems (Godwin and Batterman, 2007). Ventilation measurements across Europe show that ventilation is in practice often poor, falling in below recommended minimum levels resulting in reduced ventilation rates (lower than 0.5 ACH), increased concentrations of indoor pollutants and exposure to health risks (Godwin and Batterman. 2007; Dimitroulopoulou, 2012). Langer and Bekö (2013) similarly found that 80% of the houses they studied did not conform to the building code that requires 0.5 ACH. Similarly, a BRE study investigating the adequacy of ventilation in homes built since 1995 (when Building Regulations were revised) found that 68% of homes in the winter and 30% of homes in the summer had whole house ventilation rates below 0.5 ACH (Dimitroulopoulou, 2012).

Temporal air exchange rates (AERs) vary in commercial buildings as a result of occupancy level and behaviour (Breen et al., 2014). AER variations across residential buildings may be explained by differences in occupant behaviour and building characteristics, but also by seasonality and meteorological conditions; wind speed and outdoor temperature (Breen et al., 2014). Occupants are ambivalent when it comes to saving energy (reducing heat losses during winter and preserving coolness in summer) (Sundell et al., 2011; O'Leary et al., 2019b). Dimitrouloupou (2012) found that naturally ventilated British dwellings were better ventilated in summer (70% > 0.5

ACH) than in winter (68% < 0.5 ACH), as expected, showing that occupant behaviour (window opening) affects whole building ventilation. Liu et al. (2018) similarly found that the number of window and door openings was the most important first-order predictor of residential AER. In terms of meteorology, in summer, temperature differences and open windows increase AER. In winter, large indoor-outdoor temperature differences and high wind speeds can be equally effective in increasing AER (Breen et al., 2014). AERs are lower in other seasons as windows are closed, and the driving forces (primarily the temperature difference) are small (Breen et al., 2014; Chin et al., 2014).

Since ventilation practices vary between seasons, there is a consequential effect on indoor pollutant concentrations. During the AIRMEX study, there was a general increase in VOC concentrations (dependent on specific VOC, emission rate and building type) in the cold (winter) season owing to lower ventilation and air exchange rates (Geiss et al., 2011). For terpenes, the lowest indoor concentrations were measured during warmer seasons owing to higher ventilation rates and reactions with ozone from outdoor air, which is more abundant in warmer periods (Geiss et al., 2011). Missia et al. (2010) similarly observed an increase in pollutant concentrations in winter in response to indoor pollutant-generating activities and building materials and furnishings, as a consequence of the increased air tightness of buildings. Mandin et al. (2017) similarly indicated higher concentrations of some pollutants in summer e.g., formaldehyde and ozone and others in winter e.g., benzene, α-pinene, and nitrogen dioxide owing to differential abundance of some pollutants due to seasonality and increasing building airtightness over winter.

2.5.3.1. Exhaust Ventilation and Range Hoods

In addition to considering whole-house or building ventilation, localised ventilation systems are important to limit pollutant transport through local exhausting of air. Workplaces can benefit from local exhaust, for example, in spaces with copies and printers, preventing pollutant transport and dispersion (Spengler and Chen, 2000). The use of local exhaust fans in bathrooms and range hoods above cooking appliances represent practical illustrations of efficient ventilation (Nazaroff, 2013; Lunden et al., 2015) which can remove contaminants at the source and limit their dispersal (Rim et al., 2012; Levasseur et al., 2017).

Sometimes source control is not feasible. Reducing or eliminating the processes involved in cooking in order to improve IAQ is unrealistic since cooking is necessary for the safety and enhancement of quality of a substantial number of food products (Hager and Morawicki, 2013). Houses are often too airtight to dilute pollutants from cooking by infiltration (O'Leary et al., 2019b). Studies by Vasile et al. (2016) and O'Leary et al. (2019a) have highlighted high concentrations of CO and CO_2 and high source strengths of UFP and $PM_{2.5}$ due to cooking without adequate ventilation, with the potential to negatively affect occupant health. Devices designed to remove cooking-related contaminants include range or exhaust hoods/fans (which may be mounted above the cooktop, in a kitchen wall or ceiling) and venting ovens (Singer and Delp, 2012). Kitchen exhaust fans reduce cooking related contaminant concentrations by removing emissions directly at the stove before they mix into the surrounding air and by increasing overall air exchange in the home to remove pollutants from the indoor environment (Dobbin et al., 2018).

The efficiency of exhaust fans to capture cooking-related pollutants can vary widely given consideration to a number of factors; equipment type and design, configuration,

size and location, exhaust flow rate, house geometry and user behaviour (Dobbin et al., 2018). Singer and Delp (2012) demonstrate the importance of considering multiple performance metrics to evaluate cooking exhaust hood performance including airflow, loudness, power consumption and effectiveness at removing contaminants before they mix throughout the home, the 'capture efficiency' (CE) (Singer and Delp, 2012). Capture efficiency is seen to be a better metric than airflow alone to evaluate range hood performance (Kim et al., 2018) which is a function of fan design, installed configuration, burner position and fan speed setting (Rim et al., 2012). For a given device, higher airflow generally leads to higher CE (Kim et al., 2018), though the effect varies with particle size. At the same exhaust flow rate, particle reduction is less effective for smaller particles, likely due to molecular and turbulent diffusion (Rim et al., 2012).

Experimental and simulation studies show range hoods mounted over the cooktop are essential to use during cooking to maintain good IAQ by extracting pollutants at their source before they mix into the general air of the kitchen and home (Rim et al., 2012; Logue and Singer, 2014; Lunden et al., 2015; Dobbin et al., 2018). A device that does not cover the in-use burners suffers a large penalty in CE, increasing the quantity of pollutants released into the room or residence during cooking and increasing rates of secondary pollutant formation, leading to higher concentrations throughout the post-cooking period (Singer et al., 2011; Kim et al., 2012; O'Leary et al., 2019a). When meals were prepared whilst using an extracting cooker hood located immediately over the burner particle reductions have reached >90% in each instance (Singer et al., 2011; O'Leary et al., 2019a). Hager and Morawska (2013) observed greater removal of UFP from the back burner than the front burner. Lunden et al. (2015) confirms this, with capture efficiencies of 70–99% and 4–39% for back burners and front burners respectively.

The higher rate of air exchange introduced by a fan leads to reductions in concentration. Lower exhaust flow rates can lead to elevated indoor pollution levels (Rim et al., 2012). When using an intermittent ventilation strategy, continuing to ventilate using an exhaust fan for a period of time after cooking has a significant effect on pollutant concentrations (Dobbin et al., 2018). O'Leary et al. (2019b) showed that continuing to ventilate with a cooker hood for 10 minutes after cooking has a significant effect. Choosing to continue ventilation for 10 minutes after cooking is a balance between maximising the rate of concentration reduction and psycho-social factors, such as memory and noise (O'Leary et al, 2019b). However, continuing to ventilate has been seen to have a relatively little effect on integrated exposures compared to the effects of fan flow rate and the specific fan used (Dobbin et al., 2018). For $PM_{2.5}$, the effect of running an exhaust fan for 15 minutes after cooking was similar in magnitude to the impact of a 100 cfm increase in the flow rate used while cooking (Dobbin et al., 2018).

Ventilation requirements for kitchens vary around the world (O'Leary et al., 2019a). Several building codes require that a range hood be installed in new homes to control cooking-related pollutants, and specify required airflow rates (Kim et al., 2018). In Europe, legislation addresses fan energy with minimum requirements and a labelling system for exhaust hood energy efficiency (Jacobs et al., 2016). In the UK, under the English Building Regulations and statutory Approved Document F, kitchens in new dwellings need an intermittent extract rate of 60 l/s or 30 l/s through a cooker hood, however there is no requirement to modify ventilation in existing dwellings (O'Leary et al., 2019b). Rates were chosen to remove moisture with the expectation they will dilute NO_2 and CO emitted by gas cooking however $PM_{2.5}$ and other pollutants were not considered (O'Leary et al., 2019b). O'Leary et al. (2019b) find ventilation strategies

prescribed by English Building Regulations and ASHRAE 62.2 are adequate for <12% and 75% of houses respectively when applied during cooking.

Regular and appropriate intermittent use of a kitchen exhaust fan during cooking can reduce pollutant exposure, however, decisions about their design and use requires consideration of IAQ and energy costs (Rim et al., 2012). Using extract ventilation during cooking is especially important in airtight dwellings and during the heating season when occupants reduce ventilation rates for thermal comfort and to minimise fuel heating costs (O'Leary et al., 2019a). Increasing range hood use will impact the residential energy demand though Logue and Singer (2014) showed this increase would be negligible on the total site energy. Oversized exhaust fans and over-use can significantly increase energy consumption (Rim et al., 2012). Further work needs to estimate how mechanical ventilation will affect energy demand (O'Leary et al., 2019a).

2.5.4. Air Cleaning

Where outdoor air is contaminated, or the measures outlined above are insufficient, air cleaning using filtration techniques (including electrostatic precipitation, adsorption and excitation/acceleration) have proven effective in removing contaminants originating in indoor and outdoor environments (Levin, 1991; Shaw et al., 2005; Levasseur et al., 2017). Air cleaning and filtering devices have been increasingly used in HVAC components (Singer et al., 2016; Fazil et al., 2019) whilst portable air cleaners that clean contaminated air in rooms, are especially important for vulnerable individuals (EPA, 2017b; Lavesseur et al., 2017). Air cleaning technology is important, especially when building ventilation rates are lowered to conserve energy (Zhang et al., 2011).

Mechanical air filters remove particles by capturing them on filter material whereas electronic air cleaners such as electrostatic precipitators (ESP) use electrostatic

attraction to trap charged particles (Wallace et al., 2004; EPA, 2017b). Whereas a central fan is seen to reduce particle concentrations by 25–50%, use of an in-duct ESP can reduce particle concentrations by 55–85% compared to off-fan conditions (Howard-Reed et al., 2003). The efficiency of a particle removal air filter is measured by the minimum efficiency reporting value (MERV) developed by ASHRAE (EPA, 2017b). Fazil et al. (2019) evaluated particle air filters used in central residential forced-air systems for their removal efficiencies, revealing filters with the same ratings from different manufacturers had different efficiencies for $PM_{2.5}$ and UFPs. HEPA (High-efficiency particulate air filters) have been installed in many office, laboratory and hospital buildings and clean rooms (Shaw et al., 2005).

Gas phase air filters remove gases and odours by using a sorbent (EPA, 2017b). Some of these cleaners have the potential to generate submicron particles indoors owing to reactions between ozone and VOCs. Recently phytoremediation has been proposed as an efficient and cost-effective way to remove toxins from air (Lui et al., 2007).

2.5.5. Conflicts with Energy Efficiency

Buildings consume a significant fraction of final energy consumption worldwide and are responsible for much of the anthropogenic carbon dioxide emitted that contributes to climate change (Thomsen et al., 2016). Understanding building energy performance is important in design and retrofit (Marshall et al., 2017). It is well established that ventilation represents a significant proportion (30–60%) of total energy used in mechanically ventilated buildings, and space heating dominates energy use in the home (> 60%) (Cao et al., 2016; Marshall et al., 2017). The need to reduce energy use, driven by rising energy costs and the desire to eliminate dependence on fossil fuels has become a global and national priority (Frey et al., 2014). Energy efficiency measures in

buildings focus on reducing heating and cooling loads through improving the thermal integrity of the envelope, increasing efficiency of heating and cooling equipment and reducing system energy use (Persily and Emmerich, 2012). Public policies address decarbonisation through improving airtightness and promoting energy efficient buildings (Persily and Emmerich, 2012) including the "Energy Performance Building Directive" (EPBD) which requires all new buildings to be nearly zero-energy by 2020 (Thomsen et al., 2016; Hamilton et al., 2013; Derbez et al., 2017). The inadequate thermal performance and energy efficiency of existing buildings poses a huge challenge (Vasile et al., 2016) and to meet energy efficiency targets the energy performance of nearly all dwellings needs be improved by 2030 (Hamilton et al., 2015).

Many organisations are struggling to deal with reducing energy use (lowering ventilation) and maintaining acceptable indoor air quality (IAQ) (increasing ventilation) (Spengler and Chen, 2000; Seppänen, 2008). Increased airtightness of building envelopes to reduce air infiltration or natural and mechanical ventilation rates saves energy but worsens IAQ since it will increase indoor contaminant concentrations for contaminants with indoor sources (Seppänen, 2008; Persily and Emmerich, 2012; Langer et al., 2015; Hamilton et al., 2017; Awbi, 2017). It is not desirable to increase infiltration to improve IAQ since it is associated with increased energy demand (O'Leary et al., 2019b; Dimitroulopoulou, 2012). Increasing ventilation rates to 25 l/s per person has been seen to increase energy costs (if heat recovery systems are not used) and building running costs (Wargocki et al., 2002). Strategies that improve IAQ with no significant energy impacts or that also improve energy efficiency have been considered in the literature, driven by priorities to reduce building energy consumption, including reducing contaminants at the source, improving ventilation and purifying the indoor environment (Lavasseur et al., 2017).

2.5.5.1. Smart and Lower Ventilation

Ventilation makes up a large proportion of the energy consumption in buildings (Guyot et al., 2018) and is an attractive target for energy saving. More efficient ventilation systems are the focus of strategies to improve IAQ and energy efficiency. Natural ventilation (NV) has the potential to save fan electrical energy and NV rates can be much higher than mechanical ventilation (MV) (Schulze and Eicker, 2013) however there may be problems with ventilation heat loss. To reduce energy penalties in MV it is necessary to improve pollutant removal performance without increasing air flows and ventilation rate (Singer and Delp, 2012). To better address energy and IAQ issues, ventilation needs to be smarter. A key smart ventilation concept is to promote higher ventilation rates at times when it provides an energy and/or IAQ advantage and lower ventilation rates when it provides an energy and/or IAQ disadvantage (Guyot et al., 2018). It is favourable to include smart ventilation strategies in standards. European buildings with low energy consumption can have lower rates of building related health symptoms indicating the importance of proper design, installation and qualified, well trained operational personnel who understand the requirements for good IAQ and energy efficiency (Seppänen, 2008).

Reducing ventilation rates has negative impacts on IAQ, as such ventilation can be better controlled by sensible temperature-based-air-side economizers, enthalpy-based-air-side economizers and demand controlled ventilation (DCV), which are demonstrated in many buildings (Chao and Hu, 2004). DCV, a smart ventilation strategy, has been considered in the literature as a cost effective, energy efficient measure that also promotes good IAQ (Guyot et al., 2018). These systems adjust outside ventilation air based on the number of occupants and their ventilation demands (Guyot

et al., 2018). Traditional DCV systems use CO_2 sensors to measure occupancy as it is seen as a good surrogate for occupant-related contaminant concentrations, however these systems only guarantee that fresh air intake is sufficient to dilute these pollutants (Chao and Hu, 2004). Chao and Hu (2004) overcame the issue of reducing non-occupant related contaminants by developing a dual-mode DCV system targeting buildings where the number of occupants varies frequently. CO_2 and radon are used for sensor control to indicate the demand for fresh air to dilute non-occupant related indoor contaminants (Chao and Hu, 2004). Acceptable IAQ can be achieved using this dual-mode system and when compared to fixed-rate ventilation 8.3–28.3% of the daily electrical energy was saved (Chao and Hu, 2004).

Hesaraki and Holmberg (2015) highlight the consequences on IAQ and energy when using DCV in new housing. Results indicated that when reducing ventilation rates for the entire period of un-occupancy, VOC concentrations were unacceptable, so it was suggested that they were increased to normal requirements 2 hours before occupancy (Hesaraki and Holmberg, 2015). However, it should be noted that VOC concentrations were expected to be higher in this new building (Hesaraki and Holmberg, 2015). Laverge et al. (2011) found DCV strategies that combined manipulation of supply, vent and exhaust fan had an energy saving potential of 60% (Laverge et al., 2011).

2.5.5.2. Energy-Related Building and Retrofits

An increase in building energy performance in the EU is important to alleviate energy import and comply with the Kyoto Protocol and European Directive (2002/91/EC) on the EPBD (2018/344/EU) (Poel et al., 2007; Ekins and Lees, 2008; Langer and Bekö, 2013). The most significant impact of the EPBD is the requirement for buildings to have an energy performance certificate, indicating its energy performance, when sold or

rented, and for existing buildings over a certain size to upgrade their energy performance when renovated (Ekins and Lees, 2008) to influence the market towards energy efficient buildings (Marshall et al., 2017). Low-energy and passive houses have become more common in recent years which utilize numerous technologies including efficient insulation, advanced window technology, airtightness, and heat recovery techniques to significantly reduce energy consumption (Langer and Bekö, 2013).

Many countries have committed to constructing energy efficient buildings (Prasauskas et al., 2016). Similarly, in an effort to reduce energy consumption under the EPBD, many EU member states have introduced building retrofit programmes for existing buildings which involve improving airtightness of the building envelope (Prasauskas et al., 2016). If properly implemented alongside ventilation, energy retrofits in housing can improve thermal comfort and occupant satisfaction (Du et al., 2015), and improve mental health and reduce cardiorespiratory disease by reducing pollutant exposure (Hamilton et al., 2015). However, energy efficiency retrofits that increase the building airtightness may increase exposure to indoor-generated pollutants, negatively impacting on those with respiratory conditions (Hamilton et al., 2015). Brokerick et al. (2017) suggest that while an energy retrofit had benefits for occupant comfort and building temperature; concentrations of some pollutants increased following the retrofit as a result of lower building AER caused by improved building airtightness.

2.6. Summary

There has been a significant increase over the past decade in both the number of publications in indoor air science and the depth and breadth of research in this area, largely promoted by increasing awareness of the detrimental health effects attributed to poor IAQ. In this chapter, we have fully described and summarised this published

literature, highlighting the current knowledge and understanding and identifying new and upcoming research opportunities. There are significant contributions in the literature on many main themes including understanding the sources of pollutants, the ways in which they are measured and ways in which IAQ may be improved. However, there are opportunities for further research into source characterisation within and between particular micro-environments and into the dichotomy between IAQ and energy efficiency and ways these two objectives can be met harmoniously. These opportunities form the basis of the research described in this thesis.

Firstly, there is a specific need for further research into the dichotomy between energy efficiency and IAQ and how these -sometimes- conflicting objectives can be harmoniously achieved. Strategies have been discussed that focus on supporting both of these objectives or that support improvements in IAQ without compromising energy efficiency but there is room for more research in this area. This is of great importance owing to the large energy burden that is placed on ventilation and space heating. This is particularly important as national targets and policies seek to reduce energy use and dependence on fossil fuels. This also becomes crucial as the importance of ventilation is heightened in light of the current coronavirus pandemic.

For the quantification of indoor pollutant concentrations, scientific studies have deployed expensive regulation grade monitoring instruments that provide high quality measurements, however, there is a paucity of high-resolution monitoring data for a range of indoor environments. It would be beneficial to better understand how the spatial distribution of pollution varies around a property or building or across properties or buildings with varying characteristics. Low-cost sensor technology has been examined in recent papers and compared to reference-grade instrumentation. The

possibilities of deploying such technology have also been examined. It has been seen to be useful for increasing public awareness of air quality problems and a single unit with multiple sensors can provide holistic measurements of multiple pollutants at high spatial and temporal resolution. However, there are still questions regarding the reliability and accuracy of this sensor technology.

In terms of volatile organic compounds (VOCs), which are of particular interest because of their volatile and carcinogenic nature and widespread prevalence in indoor environments, whilst there has been extensive speciation undertaken in a variety of micro-environments, there has been less research on the influence of building characteristics and the activities that take place in these buildings on the presence of specific VOCs and related exposure for building occupants within educational settings in the UK. Many studies on VOC emissions have been conducted in chambers and are not transferrable to real-world situations. Testing emissions from individual products is not reliable for predicting emissions. More work could also focus on assessing the relationship between building standards and the concentration and prevalence of VOCs.

These gaps form the basis and focus of research presented in the following chapters.

3.0. Methodology and Instrumentation

This PhD was undertaken in conjunction with NAQTS who sponsored the research. NAQTS is an SME who offer state-of-the-art air quality monitoring technology and testing services. The V2000 and its predecessor (V1000) house an array of sensor technologies and regulation-grade equipment for multi-pollutant monitoring and played a large role in the data collection throughout this thesis. These units were most importantly used to investigate the pollutant response to typical household activities including cooking in domestic (Chapter 4) and specialist test facility settings (Chapter 5). This short chapter provides technical specifications for the V1000/V2000 units and refers to details of case studies (presented in the Technical Evaluation in Chapter 9) undertaken prior to data collection for each thesis chapter. It was important to understand the accuracy of the data reported by the V1000/V2000 and their ease of use prior to their deployment in real-world environments.

3.1. V1000/V2000 Technical Specification

The NAQTS V1000/2000 units measure Particle Numbers (CPC-Based), Particle Mass (only in the V2000, laser light scattering based), CO_2 (NDIR-Based) and CO, NO_2, Ozone, Ammonia (only in the V1000) and VOCs using metal oxide (MOX) sensors with additional measurements of CO and NO_2 using electrochemical (ELECT) sensors. Utilising dual technologies for key gas measurements enables improved cross sensitivity correction algorithms to be employed. The NAQTS V1000/2000 units are also fitted with Temperature, Pressure and Relative Humidity sensors coupled with 3D accelerometer and 3D gyro for mobile applications. External GPS and Noise (dBA) measurements are facilitated through the available USB ports. There are also an optional 4 thermal desorption tubes for full VOC speciation (which would involve external

analysis by GC–MS or similar technologies). External and internal views of the NAQTS V1000/2000 units are presented in Figure 3.1 and a full technical summary is provided in Tables 3.1 and 3.2 below.

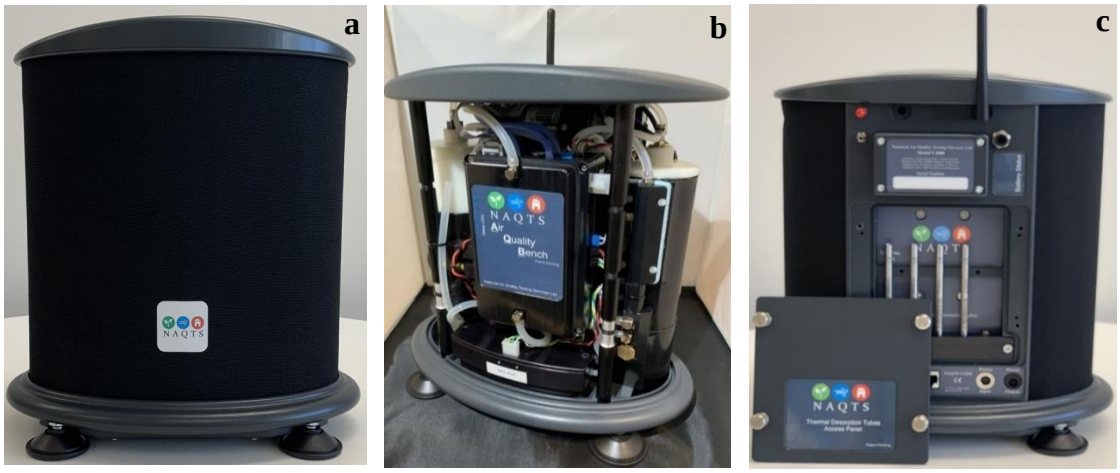

Figure 3.1: NAQTS Air Quality Monitoring Units (V1000/V2000) in cased (a) and uncased formats with (b) showing front view and (c) showing rear view.

Table 3.1: Technical specification of the V1000/V2000 units outlining capabilities and accuracies.

Particles	Specifications	Carbon Dioxide	Specifications
Technology	Mixing CPC with embedded diluter	**Technology**	NDIR
Particle Concentration Range	0–1,000,000 particles/cm^3	**Range**	0 to 5000 ppm
Concentration Accuracy	± 10% compared to reference CPC	**Accuracy**	± 30 ppm or ± 3% reading whichever is larger
Operating Temperature	0 to 30 °C	**Operating Temperature**	0 to 50 °C
Operating Humidity	0 to 95%	**Operating Humidity**	0 to 95%
Response Time	<3 secs (T10–T90)	**Response Time**	20 secs diffusion time
Working Fluid	IPA or Butanol	**Supplier**	SenseAir (K30)

Table 3.2: Technical specification continued

Environmental Measurements	Specifications
Temperature	−10 to 50 °C
Pressure	800 to 1100 hPa, ± 0.25%
Humidity	± 3% RH
Time Response	1 secs
Technology	Bosch BME-280
Power	<100 W, 12 V DC
Noise	~55dBA

Data Storage	SD Card, Local MySQL with optional Cloud Storage
Data Acquisition Rate	1 Hz
Communications	RS232, USB, Ethernet. Web-based GUI

Thermal Desorption Tubes	Use
Tenax TA	Vapor phase organics from C6/7 to C26
Graphitized Carbon	Vapor phase organics from C5/6 to C14
Tenax GR/Carbopack B	Vapor phase organics from n-C5/6 to n-C20 (EPA Methods TO-14A/TO-15/TO-17)
Tenax TA/Graphitized Carbon/Carboxen 1000	Vapor phase organics from C2/3 to C20
Carbopack C/Carbopack B/Carbosieve SIII	Vapor phase organics from n-C2/3 to n-C16/20 (EPA Methods TO-14A/TO-15/TO-17)
Supplier	Restek / Markes

We largely focused on particle number concentration (PNC) measurements provided by these units, which informed a large part of the results presented in Chapters 4 and 5 of this thesis. These measurements were provided by a condensation particle counter (CPC). A CPC counts aerosol particles by first enlarging them by using the particles as nucleation centres to create droplets in a supersaturated gas. A CPC is adjunct to an optical particle counter (OPC) that extends the range of the OPC to detect much smaller particles. The CPC housed inside the V1000/V2000 units has been calibrated by Ricardo AEA (www.ricardo.com) to provide regulatory grade measurements of particles, in line with other commercial CPC products on the market (ISO 27891). Other V1000/V2000 units used in this thesis were calibrated against the Ricardo AEA certified "gold" unit(s) to provide robust measurements with an accuracy listed above. We state that the CPC provides measurements of ultrafine particles (UFP, < 0.1 µm). We may also get measurements of particles with slightly larger diameter, but not in excess of 2.5 µm due to the construction and operation of the system. However, owing to the standard size distributions of combustion generated aerosol particles, number concentrations are expected to be dominated by the sub-100 nm range.

We also use carbon dioxide measurements (CO_2) provided by a NDIR sensor housed inside the units. This was used in some cases to determine air exchange rates (AER)

and understand occupancy levels. NDIR stands for 'Non-Dispersive InfraRed' and is a gas concentration measurement method that uses the unique adsorption wavelength range of each gas (CO_2 absorbs IR wavelength region 4.26 µm).

3.2. Case Studies

The V1000/2000 units were deployed in various indoor environments including 2 offices in Lancaster Environment Centre (LEC III), student accommodation, and a selection of residential properties (that were later used and monitored for the work undertaken in this thesis).

Table 3.3: Case studies that assess and evaluate the use and practicalities of the V1000/V2000 units and their measurement capabilities (in order of undertaking).

Date	Indoor Location	Source Type	Layered or Sequential	Pollutants Monitored
May 2017	Student Accommodation	Occupancy and Cooking	Sequential	CO_2, PNC
July 2017	NAQTS Office	Occupancy	N/A	CO_2, PNC NO_2, VOCs, CO
November 2017	LEC Office	Cleaning and Consumer Products	Sequential + Layered	VOCs
February 2018	Residence 5	Household Activities	Sequential	CO_2, PNC
May 2018	Residence 1	Household Activities and Occupancy	Layered	CO_2, PNC
May 2018	Residence 2	Household Activities	Sequential	CO_2, PNC
December 2018	Residence 2	Cooking and Log Burner	Sequential	CO_2, PNC
December 2018	Residence 4	Household Activities	Layered	CO_2, PNC

This chapter provided technical specifications for the V1000/V2000 units. The aforementioned case studies that were undertaken prior to data collection are described in detail in the Technical Evaluation (Chapter 9). Each of the proceeding thesis chapters (Chapters 4-6) involves the explicit use of or reference to these monitoring units to capture high resolution air quality information in residential and commercial environments, in large informing us about pollution from typical activities. We also see the practical use of this novel equipment in the real world.

4.0. Characterising Pollutant Response to Discrete Cooking Events and Exploring the Effects of Ventilation in Residential Environments

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Manuscript has been prepared for potential submission. This study was conceived and designed by PhD Student Charlotte Farr with extensive collaboration from my supervisory team at Lancaster University; Prof Duncan Whyatt, Dr Andrew Sweetman and Douglas Booker. The manuscript was written by Charlotte Farr, with editing and corrections made by the supervisory team.

Abstract

In the developed world, we spend most of our time indoors where we are subjected to a variety of particles mainly generated by occupants, through combustion and thermal related activities. This pilot study deploys multiple high-resolution air quality monitoring units across a number of UK residences to characterise temporal and spatial particle responses to typical episodic cooking activities, and to assess the controls on particle concentrations including natural and mechanical ventilation and housing layout.

We evaluated particle number concentration trends for different source and ventilation scenarios across eight houses and used 5 key metrics to assess critical differences between them. Results indicate that residents can be exposed to particle number concentrations up to 100 times higher than background concentrations during cooking activities, but these can effectively be reduced through natural or mechanical ventilation within a few minutes of peak concentrations being reached, with natural ventilation most effective in this respect. Results also indicate that high particle number

concentrations can persist for extended periods elsewhere in the house, depending on the layout of the house, which has implications for exposure reduction.

Key words: particulates, ultrafine particles, cooking, residential, indoor air quality, ventilation

4.1. Introduction

In the western world we spend approximately 65% of our lives in our homes, where we are subjected to various airborne particles (Klepeis et al., 2001). Indoor air quality (IAQ) is influenced by ambient concentrations, including particles associated with vehicular traffic and industrial activities which ingress into the built environment by infiltration and/or ventilation systems (Cincinelli and Martinelli, 2017). Particles also originate inside buildings from building materials and furnishings, activities undertaken within buildings and the presence and behaviour of occupants (Han et al., 2010; Kumar et al., 2016; Cincinelli and Martinelli, 2017)**.** Existing research has focused on ambient particle sources, but there has been a growing interest in risks posed by indoor particle sources as people typically spend most of their time indoors. The majority of airborne particles in residences, when expressed as particle number concentrations (PNC) are generated by the residents themselves through combustion/thermal related activities including cooking, wood-burning, candle burning and smoking (Isaxon et al., 2015; Fantke et al., 2017). Numerous studies have evaluated particle response to these activities (Hussein et al., 2006; Wallace, 2006; Wierzbicka, 2008)**.**

Cooking is seen as the most important indoor episodic activity to affect particle concentrations and is one of the most significant sources of particle emissions in homes (Dennekamp et al., 2001; Wheeler et al., 2011; Rim et al., 2012; Klein et al., 2019). Particle concentrations can reach potentially hazardous levels in the kitchen space and

throughout the building and contribute significantly to personal exposure and adversely affect health if concentrations are not maintained below health-based thresholds (Logue and Singer, 2014; Lunden et al., 2015; O'Leary et al., 2019a). The processes used in cooking such as frying, roasting, grilling, boiling and broiling contribute to particle emissions. These are also affected by ingredients, recipes and procedures, fuel types, cooking temperature and extraction/ventilation equipment (Abdullahi et al., 2013; O'Leary et al., 2019a; Klein et al., 2019).

Particles generated by combustion-related activities such as cooking are generally within the ultrafine (diameter < 0.1 µm) and fine ($PM_{2.5}$) size ranges (Abdullahi et al., 2013). The harmful effects of these particles has been reviewed in the available epidemiological literature. Due to their small size, ultrafine particles (UFP) are believed to exert higher toxicity than larger particles (Ohlwein et al., 2019). They can penetrate deeper into the respiratory system and can deposit there with a higher probability than larger particles because of their diffusion co-efficient, causing inflammatory effects (Afshari et al., 2005). UFP can also be carriers for air pollutants such as polycyclic aromatic hydrocarbons, some of which are known carcinogens. Particle Number Concentration (PNC) is the most commonly used particle metric to evaluate UFP responses. Most studies analyse particle mass and size distributions and as such data collection of UFPs is not found everywhere and epidemiology is not as solid as it is for other pollutants (e.g., $PM_{2.5}$).

Cooking has been seen to cause the highest particle concentrations in many IAQ studies and to explain most of the variation in exposure among houses (Bhanger et al., 2011). The influence of various processes on cooking emissions has been examined in the literature including the effect of different fuel types. Studies have consistently found

that gas stoves emit more particles than electric stoves (Buonanno et al., 2009; Dennekamp et al., 2001). Isaxon et al. (2015) took time resolved PNC measurements for 22 Swedish homes and found source strengths of cooking activities associated with toasting, boiling and frying activities to be highest, ranging from 1.6×10^{12} to 4.5×10^{12} particles per min^{-1}. Other researchers have also investigated cooking emissions and influencing factors and have found emission rates to be highly variable for single ingredients (Afshari et al., 2005; Isaxon et al., 2015; Dennekamp et al., 2001). Garrett et al. (1998) concluded that peak concentrations may be more important for health effects than long-term concentration averages. Studies have also assessed $PM_{2.5}$ emissions from complete meals (He et al., 2005; O'Leary et al., 2019a), finding $PM_{2.5}$ concentrations to be 30–90 times higher than background levels during frying and grilling. The Home Observations of Microbial and Environmental Chemistry study (HOMEChem) recently examined the influence of everyday activities on the emission, and found cooking was a large source of VOCs, CO_2, NO_x and particles, which were predominantly in the ultrafine mode (Farmer et al., 2019).

A limited number of studies have examined the effects of cooking in the kitchen on concentrations in other rooms. Hussein et al. (2006) found cooking activities produced total PNC (predominantly UFP) exceeding 1.8×10^6 particles/cm^3 in the kitchen with a lifetime of between 4–6 hours. This study highlighted that PNC in the living room were affected significantly when the living room door was opened, and to a lesser extent when it was closed (Hussein et al., 2006). Wan et al. (2011) similarly found cooking activities increased the PNC in the kitchen and living room after cooking, with PNC in the kitchen and living room about 20–40 times and 10 times the background level respectively (Wan et al., 2011).

Eliminating the processes involved in cooking in order to improve IAQ is unrealistic since cooking is necessary for the safety and enhancement of quality of a substantial number of food products (Hager and Morawicki, 2013). There are, however, ways to reduce emissions in domestic kitchens, including using different fuel sources, non-stick frying pans and cooking methods that avoid the browning or charring of food (O'Leary et al., 2019a). Effective mitigation strategies including natural ventilation and mechanical ventilation are therefore necessary to reduce exposure to particles. The former occurs through unintentional leaks in the building envelope, intentional openings such as windows and via coupled spaces such as basements (Liu et al., 2018a). Mechanical ventilation is particularly important during the heating season when occupants seek to reduce natural ventilation rates to enhance thermal comfort or minimize heating fuel costs (O'Leary et al., 2019a).

It has been found to be most effective to extract particle emissions at source using a cooker hood since the added air exchange introduced by the exhaust fan leads to reductions in concentrations (Dobbin et al., 2018). The efficiency of exhaust fans to capture cooking-related pollutants can vary widely based on a number of factors including equipment type and design, configuration, size and location, exhaust flow rate, exhaust ducting, installation details and use behaviour and house geometry (Dobbin et al., 2018). Higher range hood flow rates are generally more effective for UFP reduction, though the reduction varies with particle diameter due to molecular and turbulent diffusion (Rim et al., 2012). The ability of a cooker hood to capture particles is indicated by its capture efficiency (CE). O'Leary et al. (2019a) found that particle emissions could be reduced most significantly using a cooker hood with a CE of > 90% and a non-stick frying pan.

The extent to which the exhaust device extends over the burners being used has a large influence on CE. A device that does not cover the in-use burners suffers a large penalty in CE, increasing the quantity of pollutants released into the room during cooking, and leading to higher concentrations during the post-cooking period (Singer et al., 2011; Rim et al., 2012; Dobbin et al., 2018; O'Leary et al., 2019a). When adopting an intermittent ventilation strategy, using an exhaust fan for an extended period of time once cooking has ceased can more notably reduce pollutant concentrations (Dobbin et al., 2018). Dobbin et al. (2018) found that 15 minutes of additional fan use significantly reduced integrated exposure to UFP and $PM_{2.5}$. O'Leary et al. (2019b) showed that continuing to ventilate with a cooker hood for a further 10 minutes after cooking had a greater effect on reducing particle concentrations. However, the decision to continue to ventilate for a further 10 minutes was somewhat random, being a trade-off between maximizing the rate of concentration reduction and psycho-social factors, such as noise (O'Leary et al., 2019b).

It is important to understanding cooking-related emissions so we can assess the best and most appropriate mitigation strategies. Whilst considerable research effort has been expended on monitoring indoor particle concentrations resulting from episodic household activities, few studies have utilised multiple high-resolution monitors simultaneously in the indoor environment. Furthermore, few researchers have explored how exposure mitigation varies between properties of varying age and structure. The main aim of the study is therefore to analyse particle number concentrations (PNC) associated with a series of discrete cooking events, and the ways in which these change in response to different types of ventilation in houses with different characteristics.

The objectives of this study are therefore to;

1. Take high time-resolved measurements of PNC associated with discrete cooking events within different microenvironments for a selection of houses in NW England;
2. Develop a series of metrics that can be used to quantify differences in PNC caused by different source types and different forms of ventilation;
3. Evaluate the influence of natural and mechanical ventilation on the decay of PNC concentrations across houses of varying age and structure;
4. Explore how PNC within an individual household may be influenced by other factors including housing layout.

4.2. Methodology

4.2.1. Measurements and Instrumentation

For 7 non-consecutive days, high resolution (1-second) measurements of PNC were taken from fixed locations within 8 purposely selected houses in NW England that were accessible for monitoring. All houses were occupied during the monitoring period, but un-occupied during active periods of monitoring aside from the investigator. The characteristics of the individual houses are summarised in Table 4.1. The measurements were taken between July 2018 and April 2019.

Table 4.1: Characteristics of the residences monitored over the course of this study; type, age, hob type (Gas vs Electric), oven type (Gas vs Electric), ventilation strategy, kitchen volume.

House	Type	Age (years)	Hob	Oven	Vent	Kitchen Volume (m^3)
1	Terrace	120	Gas	Electric	A	48
2	Detached	17	Gas	Electric	B	56
3	Terrace	5	Gas	Electric	B	65
4	Semi-Detached	60	Gas	Electric	A	47
5	Detached	26	Gas	Electric	B	123
6	Semi-Detached	60	Gas	Electric	A	29
7	Flat	20	Electric	Electric	A	86
8	Terrace	120	Gas	Gas	B	15

A: Mechanical ventilation in kitchen (re-circulating) plus natural ventilation
B: Mechanical ventilation in kitchen (venting) plus natural ventilation

PNC were monitored with a condensation particle counter (CPC henceforth), housed inside an NAQTS V2000 unit. Notwithstanding the epidemiological evidence that UFPs may be more harmful to health than large particles, combustion generated particles tend to be considerably smaller than 2.5 µm. This justifies the use of PNC as a more relevant metric than mass concentration to determine residential exposure.

4.2.2. Experimental Design

Occupants were asked to complete a structured questionnaire prior to monitoring. This was used to capture information on construction year, floor and wall materials, and ventilation systems of the individual houses. Floor plans were provided by the occupants where available. A laser distance meter was used to measure kitchen area and volume.

PNC were recorded for a series of discrete cooking events (Table 4.2). The cooking activities included toasting bread in a toaster, frying an egg on a hob, and cooking bacon in an oven. The ingredients were selected because they are typically used across most UK households and the activities could easily be replicated. During toasting, two pieces of bread were cooked on the highest toaster setting for 5 minutes. This process is simple and repeatable with fewer variables than many other cooking processes (O'Leary and Jones, 2017). During frying, an egg was fried on a hob with a small amount of olive oil for 6 minutes after heating the oil in a frying pan for 2 minutes. During oven cooking, three rashers of bacon were cooked at 200 °C for 10 minutes after pre-heating the oven for 10 minutes. Each activity was performed under two ventilation scenarios (no ventilation, natural ventilation) and frying and oven-cooking activities were also

performed under mechanical ventilation. To reproduce real-world conditions, in naturally ventilated scenarios, windows were opened once cooking was complete, whilst mechanical ventilation was activated prior to the onset of cooking.

During the initial phase of experimentation two NAQTS V2000 (hereafter V2000) units were independently placed in the kitchen to investigate PNC whilst internal doors remain closed (this was not feasible in an open plan property). One unit was placed close to the cooking source and one unit was placed at the other end of the kitchen. During the second phase of experimentation, additional V2000 units were placed around the house. V2000 units placed in the same room as the cooking sources (kitchen) are herein referred to as 'near-field' monitors. Far-field monitors were typically placed in an upstairs bedroom or stairway (or in house 7 in a secondary room) with internal doors left open to promote air flow around the whole house.

Table 4.2: Episodic cooking experiments (that include toasting, frying and cooking bacon) conducted within each house under various ventilation scenarios and within one room (kitchen) and around the house (whole house). Experiment, ventilation characteristics and locality indicated.

Expt	Location	Internal Doors	Source	Ventilation	Ventilation Operated From
A	Kitchen	Closed	Toast	None	
B	Kitchen	Closed	Toast	Natural	Opened after episodic cooking
C	Kitchen	Closed	Fried Egg	None	
D	Kitchen	Closed	Fried Egg	Natural	Opened after episodic cooking
E	Kitchen	Closed	Fried Egg	Mechanical	Beginning of frying
F	Kitchen	Closed	Bacon	None	
G	Kitchen	Closed	Bacon	Natural	Opened after episodic cooking
H	Kitchen	Closed	Bacon	Mechanical	Start of pre-heating
A	Whole House	Open	Toast	None	
B	Whole House	Open	Toast	Natural	Opened after episodic cooking
C	Whole House	Open	Fried Egg	None	
D	Whole House	Open	Fried Egg	Natural	Opened after episodic cooking
E	Whole House	Open	Fried Egg	Mechanical	Beginning of frying
F	Whole house	Open	Bacon	None	
G	Whole House	Open	Bacon	Natural	Opened after episodic cooking
H	Whole House	Open	Bacon	Mechanical	Start of pre-heating

Before each experiment, the pans and cooking utensils were cleaned in warm water with standard dishwashing soap, rinsed with tap water and dried. At the end of each cooking event all cooking appliances were turned off and the frying pans/baking trays were moved aside to reduce continued emissions and to give a clear end to the experiment. In addition, after each test, once PNC had declined to background levels, each house was 'flushed' through an extended period of natural ventilation prior to the next experiment being conducted.

4.2.3. Data Processing and Analysis

All PNC profiles were visually assessed prior to further analysis. A series of key metrics were adopted to enable comparisons to be made between different source and ventilation combinations across the various houses. These metrics are similar to those used in hydrology; namely time to peak (TTP), peak concentration (PKC), time to background (TTB), rate of decay (RTE) and area-under-curve (AUC). These are illustrated in Figure 4.1. TTP and TTB are self-explanatory, relative to the timing of the peak concentration. RTE was estimated for each experiment where there was a clear rise and fall in particle numbers observed, by using a linear regression of the natural logarithm (Dobbin et al., 2018). The area-under-the-curve (AUC) is representative of the integral between two points in time, namely the start of a cooking event and the time when PNC returned to background levels. The sum of the area-under-the-curve is therefore a surrogate to source strength. An emission rate is the source strength divided by the time (duration) of cooking activity. Source strength and emission rates have been calculated in a similar manner to O'Leary et al. (2019a).

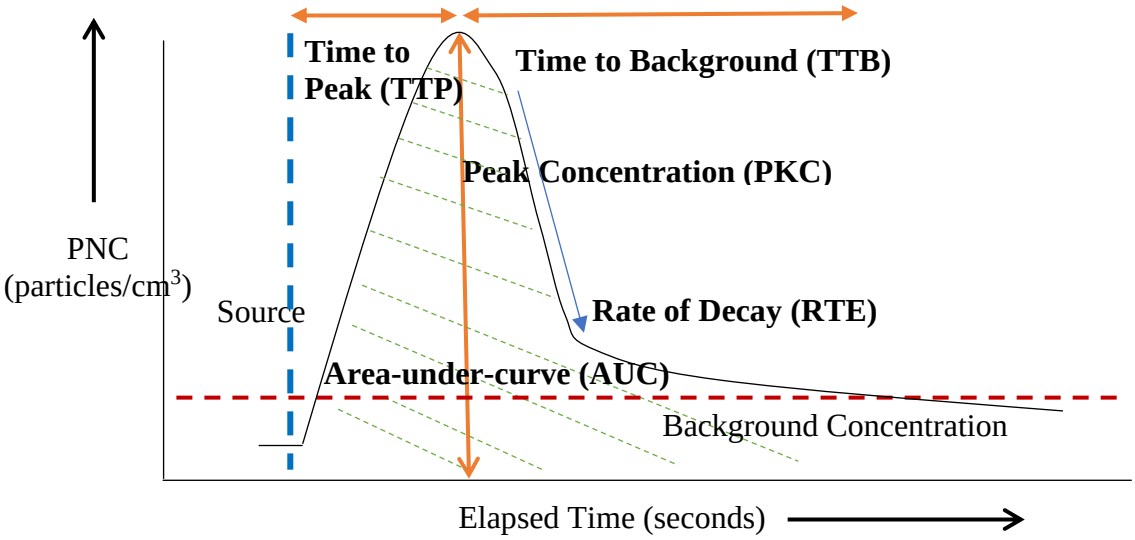

Figure 4.1: Schematic of PNC curve over time generated by a discrete cooking activity with the following key metrics highlighted; time to peak (TTP), time to background (TTB), peak concentration (PKC), rate of decay (RTE) and area-under-the-curve (AUC).

Air exchange rate (AER) calculations were based upon the slope of the logarithmic decay of PNC within a boundary where the r-squared is above 0.95 (Table 4.3). This was more reliable than the CO_2 method due the consistency in nature of exponential decay. We calculated AER under different ventilation scenarios to determine the likely dominant PN removal processes. Under no ventilation, we see a low AER (Table 4.3) and assume that particles are mainly removed by natural infiltration, more so in older buildings than new ones, and deposition. For natural and mechanical ventilation, we see higher AER (Table 4.3) and assume dispersion to be the dominant process. Houses 1, 4, 6 and 8 are older and seem to be more "leaky".

Deposition rates are considered; these are simply modelled based on the work of He et al. (2005) and also use AER calculations as a way to infer relative influences of deposition (to ventilation). We also gain some understanding of the influence of decay by subtracting the ventilated AUC from the non-ventilated AUC which eliminates the influence of ventilation.

Table 4.3: Representative Air Exchange Rate (AER) based on logarithmic decay of PNC, from oven cooking (ACH h^{-1})

House	No Ventilation	Mechanical Ventilation	Natural Ventilation
1	0.72	0.72	2.16
2	0.36	1.44	3.24
3	0.36	1.08	4.32
4	1.08	1.08	5.40
5	0.72	4.32	8.64
6	1.44	2.88	9.72
7	0.72	1.08	6.84
8	1.80	2.52	5.04
Mean	0.90	1.89	5.67

The raw data were compiled into summary tables for each metric. These tables were then imported into IBM SPSS Statistics version 26 to look at differences between means per house (for all source types and ventilation scenarios) and means per scenario (for all houses and source types). Before conducting any statistical tests the distribution of each group of data was tested to see if it was normally distributed using the Shapiro Wilks test. If the significance (*p*) value was < 0.05 then the data were significantly different from the normal distribution and the nonparametric Mann Whitney U test for two or more independent samples was used to determine whether the means were different. If the significance (*p*) value was > 0.05 then the data were normally distributed, and the parametric Independent Samples T-test was used to determine whether means were different. In both cases, a significance (*p*) value of 0.05 was used to determine whether tested means - e.g., for a ventilated and unventilated scenario - were significantly different.

4.3. Results and Interpretation

4.3.1. Characteristics of Cooking Emissions

A total of 128 discrete cooking events were characterised. These show large enhancements of UFPs (indicated by PNC) which can persist within the kitchen and

elsewhere around the house for significant periods of time. During cooking, particles can originate from both the heat source and the food, leading to some distinguishing source-specific characteristics. However, all the PNC time-series show similar profiles over time (Figure 4.2) and can be divided into three distinct periods; [1] initial background period, [2] cooking activity period, and [3] post-cooking period of decline to background, similar to periods described by Zhang et al. (2010). The PNC increases rapidly with the onset of cooking then decays at rates mainly determined by air exchange and deposition onto interior surfaces. We focus primarily on air exchange in our exploration of source-ventilation-house specific influences (Figure 4.3) however, we also consider the influence of deposition through a basic model.

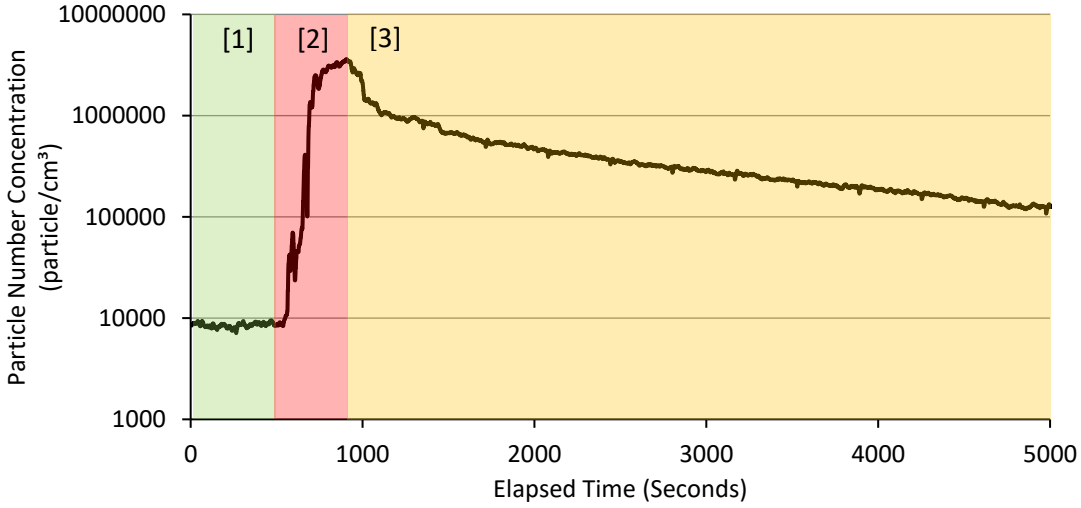

Figure 4.2: Typical temporal PNC response over an episodic period of cooking. Data taken from House 3 based on egg frying with no ventilation but indicative of PNC response across all experiments. Green [1] Background concentrations, with absence of activity, and overall good IAQ. Activity then begins at the boundary of the green and red sections. Red [2] Greatly enhanced concentrations, with cooking activities and worsening IAQ. Orange [3] Decaying concentrations, following cooking activities with improving IAQ.

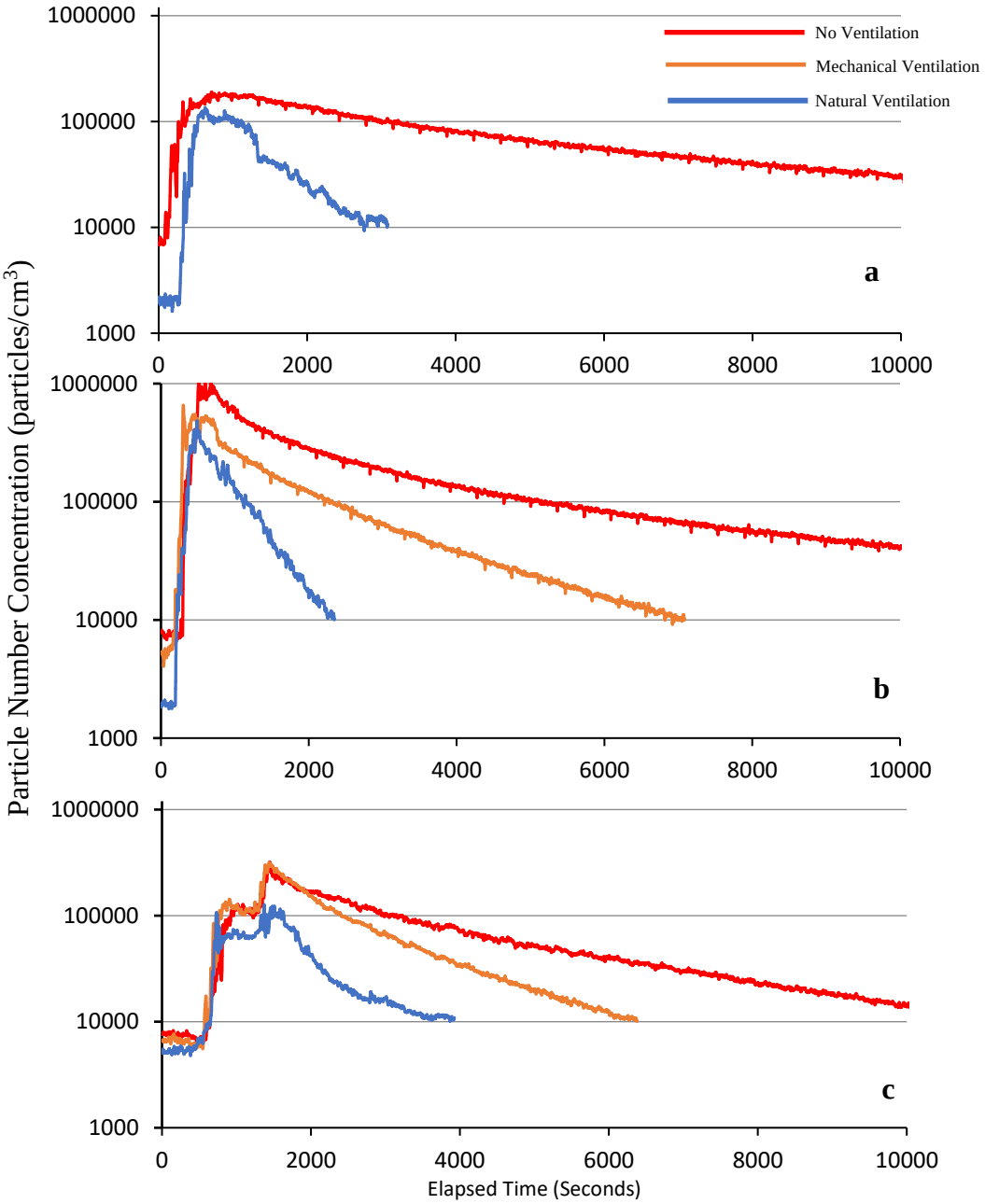

Figure 4.3: PNC response to (a) toasting bread (b) frying eggs and (c) oven cooking bacon under different ventilation scenarios. Data from near-field monitor in House 2.

Across all houses, prior to cooking, PNC typically varied between 1000 and 10,000 particles/cm^3 (often below 5,000 particles/cm^3). With the onset of cooking, PNC remained low for a short period of time (on average ~5 mins), then rose rapidly until peak concentrations were reached. PNC were typically > 10^5 higher than background

levels during cooking activities with the highest PNC higher than those reported outdoors in the world's most polluted cities (de Jesus et al. 2019). However, peak levels were short lived (on average ~5 mins). Our results reveal that peak concentration varies considerably with source type, ventilation type, placement of monitor (near and far-field), and type of housing. Emission rates varied over time as cooking continued, which was the result of a range of factors (O'Leary et al., 2019b).

Once cooking has stopped, PNC decay towards background levels. The increase in PNC immediately after the onset of cooking is typically more rapid than the decrease in PNC once cooking has ceased with the rate of decay being governed by dispersion (sometimes promoted by ventilation), deposition, and in some (high temperature) cases, coagulation. Deposition to indoor surfaces is a key sink, though much less of a significant influence than ventilation, which we know from differential air change rates between ventilated and non-ventilated scenarios (Table 4.4) as well as particle loss calculations (Table 4.5). Liu et al. (2018b) have recently established that deposition rate is linearly correlated with natural ventilation rate, but we, like many other researchers, assume a constant rate of deposition in our calculations, and expect deposition will stay roughly constant between tests for any individual house (Dobbin et al., 2018).

Dispersion promoted by air exchange and ventilation is therefore likely the most dominant process for reducing PNC, and the biggest cause for variability between scenarios and houses. In general, dispersion promoted by natural or mechanical extract ventilation ensured that particles were only present in high numbers for relatively short periods of time in all houses included in this study (~30 minutes). Using a simple model, we can quantify the significance of ventilation as a removal process (Table 4.5),

showing a much more rapid reduction in particles from an example initial peak of 1,000,000 particles/cm^3.

Table 4.4: Natural decay rates (h^{-1}) for non-ventilated and ventilated toasted scenarios. Percentage (%) of particles lost after an hour in each ventilation situation for each house. Both of which indicate the efficiency of ventilation and a reflection of the contributions of deposition and ventilation. Highlighted rows did not have strong correlation rate for calculations.

	Non ventilated		Ventilated	
House	Decay Rate (h^{-1})	% particles lost after hour	Decay Rate (h^{-1})	% particles lost after hour
1				
2	0.72	51.3	3.96	98.1
3	1.08	66.0	4.32	98.7
4	1.8	83.5	7.56	99.9
5	1.08	66.0	2.88	94.4
6	2.52	92.0	10.08	99.9
7			4.32	98.7
8	3.6	97.3	9.72	99.9

Table 4.5: Modelled particle concentrations given an example starting or peak concentration of 1,000,000 particles/cm^3 overtime highlighting the significance of the greater air change rate given during ventilation that removes many more particles over shorter time scales (given to closest thousand).

	Model Particle Concentration (particles/cm^3)	
Hour	Non-ventilation	Ventilation
0	1000000	100000
1	165000	0
2	27000	0
3	5000	0
4	1000	0
5	0	0
6	0	0

4.3.2. Evaluation of Metrics and Ventilation Measures

We have applied statistical tests of difference to compare and contrast our metrics for the different source and ventilation scenarios (summarised in Table 4.2) for the 8 houses

(summarised in Table 4.1). Summary statistics (minimum, maximum, mean, standard deviation) were generated for data captured by i) near and far-field monitors located in a single room (kitchen) and ii) near and far-field monitors located in two rooms (kitchen and one other room). We used SPSS to determine statistically significant differences between mean values for the hypotheses we were testing. We also used descriptive statistics to isolate unusual outcomes which we explored in more detail.

4.3.2.1. Near and Far-Field

Single Room (Kitchen)

Here we aimed to test whether there was a statistically significant difference between the mean value per metric for each house (based on all source type and ventilation scenarios) based on data from near- and far-field monitors deployed in the same room (with internal doors closed where possible to restrict particle movements elsewhere around the house). These tests were used to determine whether 2 units located in the same room monitored similar PNC at similar times. Table 4.6 summarises peak concentrations per house for each source type and each ventilation scenario based on data derived from the 2 units located in the kitchen. Summaries of the other metrics are illustrated in Appendix A1.

Table 4.6: Peak particle number concentrations (PKC) across all activities (toasting, frying, oven-cooking) and ventilation scenarios (no ventilation, natural ventilation and mechanical ventilation) and houses for one room at (proximal [distal]) locations (particles/cm^3 x 10^5).

	H1	H2	H3	H4	H5	H6	H7	H8
Toast No Ventilation	2.6 [3.6]	1.9 [2.8]	0.4 [3.3]	10.9 [5.4]	1.3 [2.0]	6.0 [10.6]	4.0 [9.5]	10.8 [21.7]
Toast Natural Ventilation	1.9 [4.0]	1.3 [1.6]	0.8 [1.4]	13.0 [2.8]	0.7 [1.4]	6.2 [6.1]	2.3 [7.8]	11.4 [20.3]
Eggs No Ventilation	0.8 [0.5]	10.7 [5.8]	29.2 [4.7]	19.8 [9.1]	2.3 [6.1]	15.5 [11.9]	3.9 [7.2]	16.2 [25.8]
Eggs Mechanical Ventilation	1.9 [1.3]	6.6 [3.8]	23.2 [6.4]	25.1 [12.9]	1.2 [3.8]	11.3 [8.47]	1.3 [3.7]	16.8 [22.4]

Eggs Natural Ventilation	1.6 [1.0]	4.8 [4.8]	15.8 [5.8]	22.5 [8.4]	2.0 [5.9]	12.1 [10.6]	3.2 [8.2]	16.5 [26.3]
Bacon No Ventilation	12.7 [6.1]	3.2 [2.8]	18.5 [3.7]	7.6 [2.4]	0.9 [2.3]	6.1 [3.6]	22.6 [2.2]	31.8 [18.1]
Bacon Mechanical Ventilation	16.5 [8.9]	3.2 [1.9]	14.1 [5.2]	11.5 [6.5]	1.9 [1.2]	6.1 [1.4]	16.5 [2.2]	24.5 [18.1]
Bacon Natural Ventilation	11.4 [11.3]	1.3 [0.9]	12.4 [3.0]	14.2 [3.6]	1.2 [2.24]	4.4 [1.9]	29.5 [5.0]	25.6 [17.5]
Mean	6.2 [4.6]	4.1 [3.0]	14.8 [4.2]	15.6 [6.4]	1.4 [3.1]	8.5 [6.8]	10.4 [5.7]	19.2 [21.3]

The results of statistical analyses reveal that almost all *p*-values across all metrics are > 0.05 (Table 4.7). Therefore, this informs us that although we independently measured PNC at two locations in the same room (kitchen), they generally show similar values and provide data that align with one another. We therefore conclude that generally the mean values derived from the 2 monitors were not statistically significantly different, and we broadly see the same magnitude and temporal response in PNC to our discrete cooking events in both units. Therefore, we take the measurements from the near-field monitor to be representative of the room as a whole.

Table 4.7: Summary of statistical tests to determine whether differences between the mean values derived from near- and far-field monitors were significant across all scenarios ($p < 0.05$) (p values < 0.05 highlighted in bold font) for each key metric.

House	TTP	PKC	AUC	RTE	TTB
1	0.726	0.645	1.000	1.000	0.707
2	0.467	0.402	0.645	0.435	0.895
3	0.629	**0.014**	0.145	0.959	0.804
4	0.328	**0.004**	0.321	0.442	0.952
5	**0.007**	**0.035**	**0.021**	0.645	0.534
6	0.721	0.431	0.835	0.645	0.731
7	1.000	0.878	0.721	0.878	0.898
8	1.000	0.488	0.979	0.574	0.845

We have illustrated the data for each cooking activity and calculated summary statistics and we conclude that a single V2000 monitor can provide a representative measure of IAQ in a single room. However, we also make some interesting observations and

observe some variability in individual events. For example, emissions associated with cooking bacon in an oven are always higher close to the source, whilst emissions associated with frying an egg or toasting bread are usually higher at a distance. This was particularly noticeable for House 5 and 7. This is likely a product of the way in which the food is cooked and way the pollutant plume evolves.

Multiple Rooms (Kitchen and Other Room)

Here we explored spatio-temporal variation in PNC around individual houses. In multi-room scenarios we observe high PNC throughout the property. Persistence of these particles, particularly upstairs, highlights concern with regards to health consequences since people will spend a significant amount of time here (sleeping). Here we aim to test whether there is a statistically significant difference between mean values per metric for each house (based on all source type and ventilation scenarios) for near- and far-field monitors deployed in separate rooms (with internal doors open where possible to promote particle movements elsewhere around the house) (Table 4.8). PNC data from a near-field (kitchen) and far-field (upstairs) location tells us something about the relationship between ventilation and airflow throughout a residence more generally (Appendix A2).

Table 4.8: Summary of statistical tests to determine whether differences between the mean values derived from near- and far-field monitors were significant ($p < 0.05$) (p values < 0.05 highlighted in bold font) for each key metric.

House	TTP	PKC	AUC	RTE	TTB
1	0.347	0.105	0.218	0.279	0.787
2	**0.040**	**0.000**	0.103	**0.015**	0.682
3	**0.018**	**0.002**	**0.019**	0.279	0.533
4	0.105	**0.000**	0.196	0.505	0.825
5	**0.007**	0.721	0.721	0.234	0.878
6	0.094	**0.000**	**0.005**	0.095	0.770
7	0.959	**0.021**	0.056	0.442	0.832

8	**0.038**	**0.000**	**0.025**	**0.030**	0.060

When we consider two room scenarios, we inherently increase the variability, likely as a function of floor plan and area. This variability is higher for some houses than others. We see statistical differences between mean values for TTP, PKC and to a lesser extent AUC (Table 4.8). However, no single metric shows statistically significant differences for every house, and no house shows statistically significant differences for all metrics. TTP seems moderately variable, with some houses showing significant differences between near- and far-field monitors whilst others do not. TTP is known to depend on many factors such as cooking method, particle size, relative location between the source and sampling area and indoor airflow (buoyancy and convection) (Lai and Ho, 2008). PKC differ significantly between near and far-field locations, probably because the further the particles travel before being sampled, the greater the likelihood that they will be dispersed, coagulated or deposited (Lai and Ho, 2008). This holds true for some houses in our study, but not for others, and we observe some interesting spatial patterns resulting from particle dynamics (particularly in House 1). We conclude generally that mean values for some metrics derived from the near- and far-field monitors are very different (statistically so), suggesting that the units are measuring different levels of particle pollution, though note some houses respond differently.

We used the mean value per metric across the eight houses to calculate average near-field (kitchen) and far-field (upstairs) values (Table 4.9). From TTP we see it takes almost twice as long to reach the peak concentration in the far-field. This makes sense given the extended distance from source. PKC and AUC values are much higher at the near-field monitor which is not surprising given the proximity of the source to the monitor and the potential for dispersion and deposition en-route to the far-field monitor.

The RTE is quickest in the near-field, driven by the operation of local ventilation in the near-field and initial dispersion to other regions of the house.

Table 4.9: Metrics derived from near-field (kitchen) and far-field (upstairs/elsewhere) monitors averaged across all houses.

Metric	**Near-Field**	**Far-Field**
TTP	12 mins	22 mins
PKC	1,000,000 cm^3	400,000 cm^3
AUC	700,000,000 cm^3	300,000,000 cm^3
RTE	36.30 particles cm^3/s	21.86 particles cm^3/s
TTB	79 mins	81 mins

4.3.2.2. Source-Ventilation Dynamics

Single Room (Kitchen)

Here, we aim to test whether there are statistically significant differences between the mean values for each metric for different types of ventilation (no ventilation, natural ventilation, mechanical ventilation) for all sources (toast, fried eggs, oven-cooked bacon) and all houses based on a single near-field monitor placed in the kitchen (with internal doors closed where possible to restrict particle movements elsewhere around the house) (Table 4.10). We evaluated most of the metrics we have previously used but excluded those that were not of relevance to ventilation and did not show significant differences.

Table 4.10: Summary of statistical tests to determine whether differences between the mean values derived from near- and far-field monitors were significant ($p < 0.05$) (p values < 0.05 highlighted in bold font) for relevant metrics; TTP – Time to Peak; PKC – Peak Concentration; RTE – Rate of Decay.

Source	**Ventilation**	**AUC**	**RTE**	**TTB**
Toast	NON v NAT	**0.001**	**0.000**	**0.001**
Eggs	NON v NAT	**0.016**	**0.007**	**0.001**

Eggs	NON v MEC	0.231	0.108	**0.038**
Eggs	NAT v MEC	0.168	**0.011**	**0.004**
Bacon	NON v NAT	0.065	**0.010**	**0.007**
Bacon	NON v MEC	0.721	0.173	0.279
Bacon	NAT v MEC	0.195	**0.018**	**0.002**

We see statistically significant differences in RTE and TTB but not in the other metrics (Table 4.10). Differences between TTP and PKC in near- and far-field locations are not statistically significant, which makes sense given that these metrics relate to source strength which is broadly similar across source types and ventilation scenarios. We observe a longer TTP for oven cooking in response to the prolonged cooking period (20 minutes) and hence 'mixing' duration. We tend to observe higher peak concentrations (PKC) in non-ventilated scenarios when the kitchen exhaust fan is turned off and the windows remain closed. Peak concentrations are not maintained for long periods, particularly with the onset of ventilation. Generally, for most houses, we see higher peak concentrations when oven cooking bacon ($0.86 \times 10^5 – 31.5 \times 10^5$ particle/cm^3) than for the other food types with an unusual concentration profile over time (Table 4.6; Figure 4.3), with two distinct peaks in response to opening and closing the oven door which promotes particle dispersion within the kitchen space. This is consistent with findings from the HOMEChem study where there are rapid increases in PNC and ammonia concentrations as the oven is opened which the researchers relate to the thermal decomposition of amino acids in meat proteins (Ampollini et al., 2019).

Despite not simultaneously measuring PNC outside of the residences, infiltration rates are expected to be low across the eight houses included in this study, as PNC took more than an hour on average (but sometimes significantly longer) to decay to background levels (see Appendix A1) under non-ventilated scenarios with deposition the dominant removal mechanism. This indicates that the houses are relatively airtight, despite the

age of some of the properties. Air exchange rates (AER) measured under non-ventilated conditions support this assertion (mean of 0.90 ACH per hour, Table 4.3). The main control on AER was provided by natural ventilation (window opening) or mechanical ventilation (range hood or exhaust fan) with mean AER of 5.67 and 1.89 ACH per hour, respectively. Enhanced rates of air exchange provided by natural or mechanical ventilation ensured that PNC were present in high concentrations for relatively short periods of time.

In the context of ventilation, we see significant differences in RTE and TTB as expected (Table 4.10). The decay rate (RTE) reflects the removal rate of particles. This was largely controlled and enhanced by the type of ventilation and is a key mechanism for particle removal (Zhang et al., 2010). Considering each cooking activity individually, the fastest RTE are associated with natural ventilation irrespective of source (Appendix A1). On average, across all houses, PNC reduced to background levels much more rapidly under conditions of natural ventilation than conditions of no ventilation (101 minutes more rapidly for toast and 106 minutes more rapidly for frying) due to increased rates of air exchange. From this we conclude that natural ventilation is the best strategy for reducing cooking-generated particles. Mechanical ventilation was also found to significantly reduce PNC in the kitchen. On average, across all houses, PNC reduced to background levels much more rapidly under conditions of mechanical ventilation than under conditions of no ventilation (28 minutes more rapidly for oven cooking and 54 minutes more rapidly for frying). However, these rates were highly variable across houses.

AUC shows some significant differences between near- and far-field monitors (Table 4.10) which can be related to the effectiveness of ventilation. The AUC ranged from

2.17×10^7 to 2.40×10^9 particles/cm^3. These numbers are slightly lower than those previously reported by Zhang et al. (2010). Under non-ventilated scenarios (particularly egg frying and oven cooking) we see high PNC and large AUC values. However, we cannot compare our values to health-based standards because these do not currently exist for PNC.

In summary, we do not observe significant variations in source strength. The V2000 monitor records similar PNC for toast, fried eggs and oven-cooked bacon across all houses and our statistical analysis of our source-based metrics TTP and PKC confirms this. In contrast, we do observe statistically significant differences in our ventilation-based metrics RTE, TTB and AUC. We conclude that natural ventilation is most effective at reducing potential exposures.

Multiple Rooms (Kitchen and Other Room)

Here we aim to test whether there are statistically significant differences between the mean values of each metric for different types of ventilation (no ventilation, natural ventilation, mechanical ventilation) for all sources (toast, fried eggs, oven-cooked bacon) across all houses based on a near-field monitor placed in the kitchen and a far-field monitor placed elsewhere in the house (with internal doors open where possible to promote particle movements around the house) (Figure 4.11).

Table 4.11: Summary of statistical tests to determine whether differences between the mean values derived from near- and far-field monitors were significant ($p < 0.05$) (p values < 0.05 highlighted in bold font) for each of our key metrics.

Source	Ventilation	TTP	PKC	AUC	RTE	TTB
Toast	NON	**0.041**	**0.038**	**0.029**	0.075	0.712
Toast	NAT	**0.024**	**0.001**	**0.020**	**0.027**	0.846
Eggs	NON	**0.004**	**0.015**	0.100	0.054	0.362
Eggs	NAT	**0.021**	**0.021**	0.247	0.147	0.303

Eggs	MEC	**0.032**	**0.010**	0.074	**0.007**	0.969
Bacon	NON	**0.000**	**0.004**	**0.028**	0.190	0.716
Bacon	NAT	**0.008**	**0.006**	0.141	0.529	1.000
Bacon	MEC	**0.000**	**0.006**	**0.033**	0.083	0.721

We expected the far-field monitor (typically located upstairs) to record lower PNC than the near-field monitor, and this is usually the case (Appendix A2). Differences between mean values of TTP, PKC and AUC derived from near-field and far-field monitors were generally statistically significant (less so for AUC). TTP was generally quicker at the near field monitor due to the short distance between the source and the monitor, leaving little time for removal processes. Similarly, PKC and AUC were higher at the near-field monitor for the same reason. There were also statistically significant differences between the near- and far-field monitors caused by local ventilation in the near-field only. RTE was faster in the near field in response to local ventilation. We observed variability in decay rates across all houses under naturally ventilated scenarios due to the higher rates of decay in the kitchen than elsewhere around the house. This was in part due to faster initial decay rates which includes dispersion around the kitchen and the rest of the property. This was to some extent a function of housing volume, which we later discuss. As before, we are reasonably confident in the assertion that natural ventilation is more effective at reducing PNC than mechanical ventilation in the kitchens of the eight houses we tested based on analysis of these metrics and assessment of raw data.

4.4. Discussion

4.4.1. Indoor Sources and Particle Dynamics

The factors governing indoor PNC include direct emissions from indoor sources, ventilation supply from outdoor air, filtration, deposition onto indoor surfaces, and

removal from indoor air by means of ventilation (Nazaroff, 2004). In this study, cooking is seen to be a large source of (ultrafine) particles as high concentrations were observed, as has similarly been noted in numerous other studies including HOMEChem (Farmer et al., 2019) where cooking was seen to be a large source of VOCs, CO_2, NO_2, and particles of various sizes (Farmer et al., 2019; Patel et al., 2020). Particle emissions from cooking events are intermittent, episodic and localized. The effects of emissions on inhalation exposure depend, to an extent, on indoor-air mixing processes (Nazaroff, 2004), which can be influenced by ventilation and occupancy. Following emission, the concentrations of particles indoors are the result of several processes where the production of particles is balanced by loss through various removal or transformation mechanisms (Ruzer and Harley., 2012).

Airborne particles deposit on indoor surfaces after collision and adhesion (Nazaroff, 2004; Ruzer and Harley, 2012). Surfaces therefore play important roles in the lifetime and reactivity of pollutant emissions (Farmer et al., 2019). In our study we expect variable deposition rates from house to house as a function of the varying surfaces and as a result of house volume. We suspect that larger kitchens with more surfaces for deposition could in part contribute to lower AUC values. However, on the whole, owing to the ultrafine nature of particles generated in our study by cooking activities we do not expect deposition to be a significant influence on PNC decay. We indeed attribute less than one third of particle loss to deposition, based on the variable air change rates between ventilated and non-ventilated scenarios and our particle loss calculations support this assertion (Table 4.5). Previous work also supports this conclusion. Respirable particles (diameter < 2.5 μm) such as those generated by cooking processes will remain entrained in room air movement even at higher AER (Ruzer and Harley, 2012). Indeed, settling velocities show that respirable particles (diameter < 2.5 μm) do

not deposit onto the floor quickly under the influence of gravity (Ruzer and Harley, 2012). Previous work highlights that the lowest deposition rates were found for particles in the size range from 0.2 to 0.3 μm for both minimum (AER: 0.61 ± 0.45 h^{-1}) and more typical (AER: 3.00 ± 1.23 h^{-1}) ventilation conditions (He et al., 2005). Coagulation is another potential removal mechanism (Rim et al., 2012), particularly for UFP in high concentrations. However, we were not able to include measurements of particle size in our study, with our monitor recording PNC only. Therefore, we were unable to measure the changes in size distribution over time to look at coagulation effects.

The volume of the house, the activities of its residents and methods of ventilation can have a significant effect on the concentration of indoor particles (Nasir and Colbeck, 2013). We now examine the influences of ventilation rates and house volumes on IAQ.

4.4.2. Ventilation and Air Exchange

Most houses are ventilated by a combination of natural ventilation through windows and other design openings plus infiltration and intermittent extract ventilation, including those in this study. In our study, leakage flow or infiltration appears to be low, with greater exposure to elevated PN under non-ventilated scenarios (Appendix A1 and A2) where removal is dependent upon deposition and infiltration, indicating that all houses are generally airtight (as also indicated by our low AER, Table 4.3). Therefore, infiltration is not a major mechanism for particle removal i.e., is not as effective as purpose provided ventilation. Natural ventilation, driven by wind and thermally generated pressures, seems to meet ventilation needs in this study with window opening significantly increasing the rate of air exchange in even the most airtight of houses following episodic cooking activities. Similarly, when using an extract ventilation strategy, we see significant removal of particles. We found that continuing to run the

extraction fan for the duration of PNC decay significantly accelerated the decay. We know in our experiments that the exhaust ventilation was directly over the hob so we are getting a larger reduction in particle concentration that might otherwise be expected (Singer et al., 2011; Kim et al., 2018; O'Leary et al., 2019a).

Whether the range hood is venting, or recirculating has important impacts on IAQ. Venting range hoods that exhaust kitchen contaminants directly to the outside environment (Kim et al., 2018) are most effective. Some homes in this study did not have venting systems, or the venting systems were not enabled. However, we found that even re-circulating range hoods or range hoods that did not vent properly outside (e.g., House 4) were still effective at removing particles, more so than if they were not in operation. Regardless of whether a cooker hood is venting properly or not, we observed a significantly enhanced rate of particle removal. In fact, we do not discern significant differences in pollutant removal rate irrespective of whether the cooker hood is venting outside or recirculating.

In the UK, under the English Building Regulations and Approved Document F, kitchens in new dwellings are required to have an intermittent extraction rate of 60 l/s or 30 l/s through a cooker hood (Kim et al., 2018; O'Leary et al., 2019a; UK Government, 2014a). Only one such property would have been designed with these regulations in mind (House 3). All other properties investigated in this study were built before these regulations came into place, so we cannot compare between guidelines and real-world situations. There is also the issue of maintenance and as such few extractor fans will be operating at published CE. Actual usage of intermittent extract ventilation also needs be considered. In reality, occupants use extraction fans in their homes and apartments

less frequently than might be expected. For example, Park and Kim (2012) found that only 31% of occupants used their fans in a study based in Korea.

4.4.3. Monitoring Room and House Volume

Another consideration in relation to IAQ is the floor area and room volume. We focus here on the toast source only and kitchen volume (total of 16 observations). Home or room volume has been evaluated in relation to IAQ in previous studies (e.g., Hubuyo et al., 2011). Consistent with such studies we find the smaller the volume of the room in which the monitor is placed, the higher the PNC (Figure 4.4) (Haghighat and Kim, 2009). Whilst our results show a low degree of correlation (R^2 of 0.15) between AUC and kitchen volume under non-ventilated conditions, they show a moderate correlation (R^2 of 0.51) under naturally ventilated conditions, indicating that as kitchen volume increases, AUC decreases. The non-linearity in this trend is consistent with the variation in source and ventilation between homes. It is possible that variations in source strength or more likely ventilation rate (and air exchange provided by window opening) could have some influence on this trend, however, we assert some influence driven by differences in volume.

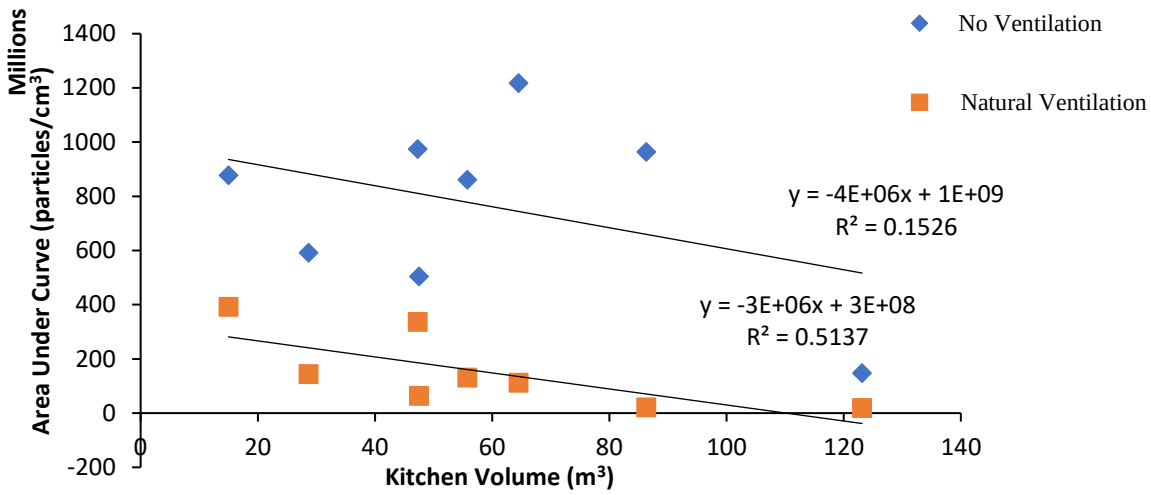

Figure 4.4: Correlations between kitchen volume and area-under-the-curve plots (which is indicative of total amount of toast-generated PNC in the room and surrogate for source strength) for each of the eight houses monitored in this study under non-ventilated and ventilated scenarios.

Klepeis et al. (2017) similarly found at lower room volumes some homes have much higher particle levels than others. A recent study of particle size in relation to building characteristics by Urso et al. (2015) likewise found lower levels of fine particles were associated with larger houses and the use of kitchen air-exhaust systems. Jetter et al. (2002) also found that particle emissions from burning incense were high in small, poorly ventilated rooms. We conclude that in our study this is because (assuming roughly similar source strengths), we see a much larger accumulation of particles in smaller rooms (per unit of air) which can be reduced to background levels much more rapidly than an equivalent number of particles in a larger room. This is also indicated by the moderate correlations between RTE and kitchen volume for non-ventilated (R^2 = 0.50) and ventilated (R^2 = 0.58) results (Figure 4.5). We attribute slower decay rates in larger kitchens to smaller source impacts (despite larger mixing volumes for diluting pollutant concentrations (Klepeis et al., 2017)) due to larger volumes of air and thus smaller concentration gradients. Slower decay rates in larger kitchens might also indicate deposition is more of a controlling factor that we previously asserted. This

contradicts the findings of Jovasevic-Stojanovic and Bartonova (2017) who found that smaller free space areas were associated with longer residence times and prolonged exposure.

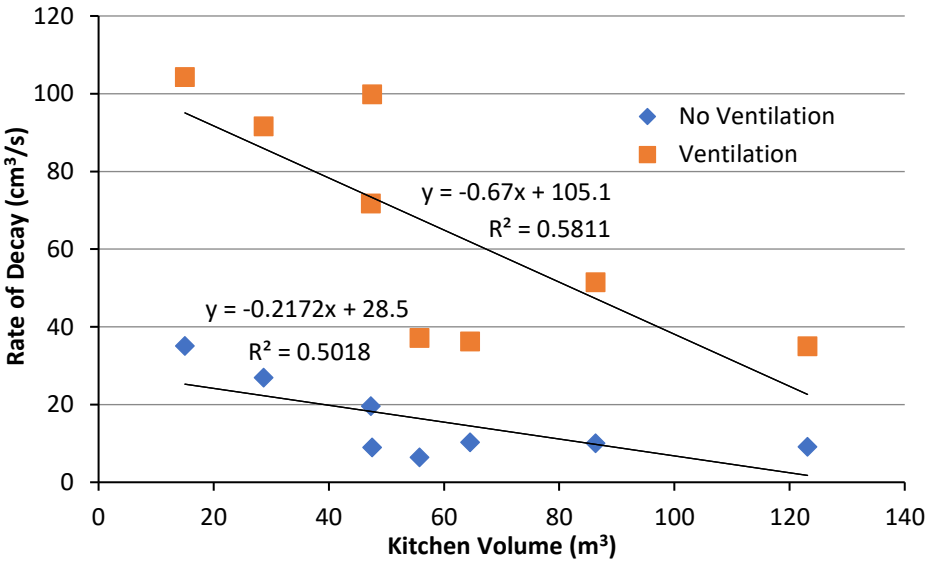

Figure 4.5: Correlation between kitchen volume and decay rate for toasting activities under non ventilated and naturally ventilated scenarios across all eight houses in this study.

The size of the room will also dictate the available surface area for deposition. We hypothesise that increasing the room volume will increase the number of surfaces available for interactions and deposition. For example, Thatcher et al. (2002) found that surface area of a bare space (nominal surface area of 35 m^2) could be increased by 12 m^2 through the addition of furniture which increased the deposition rate by a factor of 2.6. This would mean that we would expect lower particle exposures associated with larger monitoring room volumes, which is what we see in our AUC metric (Figure 4.4) but not in our RTE metric (Figure 4.5). However, since we do not quantitatively consider the influence of deposition (due to our focus on PNC measurements) and assert the influence of deposition to be negligible in comparison to ventilation, we are confident in this assertion. We take the air change to volume ratio to be important, not volume alone.

When considering whole house dynamics, we expect higher PNC in houses with smaller volumes and lower PNC in houses with higher volumes for the reasons stated earlier. Klepeis et al. (2017) found that houses with more doors, bedrooms, or bathrooms were generally bigger and that particle levels tended to be lower as a consequence of the greater mixing volume for diluting pollutants. We reach the same conclusion. The type of home also appears to influence PNC in the upper quartiles of the distributions with apartments having higher PNC than detached houses (Klepeis et al. 2017). We do not discern this effect between detached houses and apartments, but this could be attributed to our small sample size.

These results and our interpretation might not match those for other sources (eggs, bacon) or extended spaces. Correlations between kitchen volume and our metrics derived for frying eggs and oven cooking bacon are weaker (Appendix A3). These activities occur over longer periods of time and are more complicated than toasting. It is conceivable that rather than observing 'pencil-like' plumes as we did with toasting, we might see much broader spread of particulates from these activities that would evolve in a different manner. The particle size and composition are also likely to be more complex given the nature of these sources.

4.4.4. Housing Structure and Layout

When windows and internal doors were closed PNC in the kitchen remained at high levels for longer periods of time, since the limited airflow restricted the dispersion of pollutants elsewhere around the house, effectively compartmentalising the house (Zhang et al., 2010). However, our results also suggest that relatively intermittent cooking activities can have a significant effect on PNC elsewhere throughout the house and that exposure to PNC from cooking activities is not necessarily confined to the

kitchen, particularly in the case of open plan layouts or when interior doors are left open to promote wider dispersion (Nasir and Colbeck, 2013).

Layouts can vary substantially between homes (Klepeis et al., 2017): some have open plan kitchens whilst others have separate kitchens (Nasir and Colbeck, 2013). Far-field measurements (typically upstairs) confirm that particles emitted in the kitchen were easily dispersed to other rooms in houses with and without open plan layouts, most notably when interior doors are opened. Even though PNC elsewhere in houses were generally lower than those in the kitchen, they were still up to 100 times higher than those monitored during periods of no cooking activity. Therefore, the health risk from cooking emissions may be underestimated if human exposure is only considered in the kitchen. This highlights the importance of more measurements to better capture spatial distribution of pollutants indoors, to better inform IAQ models.

We find exposure to cooking-generated PNC is significant for house occupants away from the kitchen area even in those residences with a separate kitchen. This study highlights how cooking can increase PNC concentrations from background levels in both living and upstairs rooms of a house or apartment. We observed that PNC profiles were similar in the kitchen and in other rooms when interior doors were opened, with a TTP of approximately 10 minutes. We see airflow as significant in this characterisation. Airflow between rooms, driven by pressure differences, can strongly influence indoor pollutant concentrations and fates (Nazaroff, 2004). Few studies have explored concentration variability between rooms and the factors that influence them (Nazaroff, 2004). Miller and Nazaroff (2001) found that closing a door between two rooms reduced the rate of airflow between them from 60 m^3/h to 1 m^3/h. With an open doorway,

tobacco smoke particles released in one room became rapidly mixed throughout both. We observed a similar pattern from episodic cooking.

The decay rate not only represents the effects of ventilation and particle deposition, but also the combined effects of different particle removal processes due to the interaction between the kitchen and other areas of the house (Hussein et al., 2006). The kitchen itself is an open area to the hallway and rest of the house so when cooking activities occur in the kitchen and interior doors are opened, PNC are elevated throughout the house as a result of air exchange between the kitchen and whole house (Hussein et al., 2006). We often observe faster initial decay rates, particularly where some kitchens were part of an open floor plan or where there is mixing throughout the rest of the living space when interior doors are opened.

We observed that cooking emitted particles dispersed quickly from the kitchen to upstairs, indicating that potential health impacts are not limited to occupants in the kitchen. Other researchers have also found that PNC in other regions of the house were also affected when the living room door was opened (Wan et al., 2011; Hubuyo et al., 2011; Hussein et al., 2006). Wan et al. (2011) found UFPs increased by 10-fold from background levels in the living room and by 20–40-fold in the kitchen for 60 and 90 minutes respectively. Hussein et al. (2006) found the lifetime of cooking particles in the kitchen varied between 4-6 hours with a peak in the living room that was at least 30% its value in the kitchen. Similarly, Klepeis et al. (2017) identified a peak in particle concentrations in the study room was 40% of that in the kitchen. Whilst exposure to cooking emitted particles elsewhere in the house is much lower, this is not always the case for every house.

PNC measured in or near a room with an active source are expected to be higher than PNC measured in a room more distant from the source. However, we observed peak concentrations that were sometimes comparable between the near-field (kitchen) and far-field (upstairs) locations and conclude that this is a consequence of housing layout. We observed some interesting and conflicting results with regards to the influence of housing layout on trends in cooking-generated pollutants through comparisons across individual houses as we will now demonstrate with reference to Houses 1 and 2.

Generally, for most houses, lower peak concentrations and prolonged times to peak were observed in the upstairs (far-field) location relative to the near-field (kitchen) location. This is due to the relative distance between pollutant source and sampling location, and hence the distance travelled by particles, given them time to be dispersed, deposited or coagulated. This is the case for House 2. However, we observe a different trend for House 1 which appears to show different behaviour across most scenarios and metrics. Surprisingly, higher peak concentrations were recorded upstairs (far-field monitor) in House 1, which were 66–91 times higher than background concentrations (average background of ~5000 particles/cm^3) when measured 4–5 minutes after the start of the toasting events in non-ventilated and ventilated scenarios. Such rapid times to peak in House 1 are unusual for an upstairs location, and this outcome was not reproduced in any of the other houses studied. Unusually, in House 1, we also observed higher peak concentrations under the naturally ventilated scenario (4.56×10^5 particles/cm^3) than we did under the non-ventilated toasting scenario (3.34×10^5 particles/cm^3). These trends have been consistently observed in House 1 – replicated in other experiments conducted in this house prior to this study, but the geometry is particular to this house only. We would further need to repeat experiment with same source and monitor placings to confirm or otherwise refute these observations.

In terms of decay rates around the residence, we generally saw lower decay rates upstairs for both residences, which could be a response to local ventilation operating in the near field (kitchen) only. Natural ventilation reduced the period of exposure (TTB) by 1 hour 10 minutes in the near field for both houses, but 2 hours 32 minutes and 1 hour 39 minutes in the far field (upstairs) for Houses 1 and 2 respectively. In the far-field, we observed significantly longer periods of decay for House 1 irrespective of ventilation. This is attributed to enhanced peak concentrations owing to lower influence of particle removal processes (due to more proximal distance to far-field monitor) with possible influence of housing volume (smaller volume). In the absence of ventilation, the decay rates in the kitchen and upstairs were more similar.

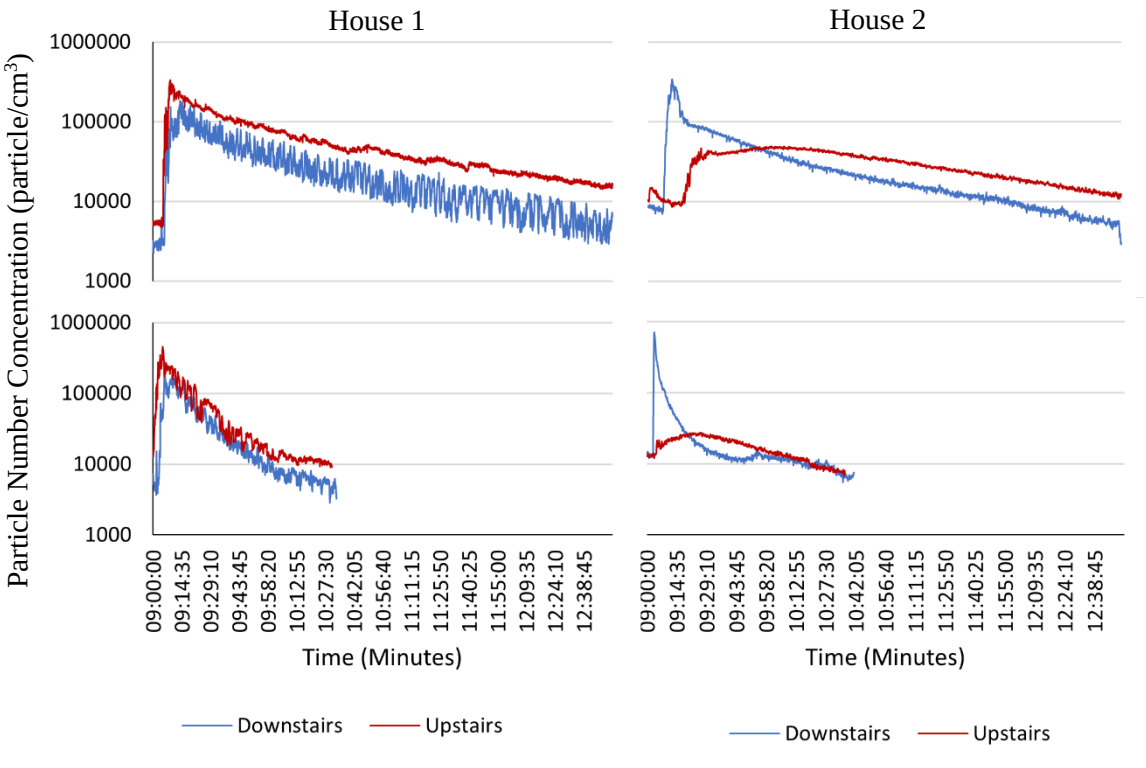

Figure 4.6: Temporal PNC trends for toasting activities for House 1 (left) and House 2 (right). Each ventilation scenario appears on a separate plot for comparison purposes. Upper plots illustrate non ventilated scenarios, lower plots illustrate ventilated scenarios.

We examine critical differences between these two properties to assess what might be responsible for some of the differences in behaviour. We find housing structure and layout to be significant factors controlling the dispersion of cooking-related particles. House 1 has an open-plan kitchen with the toaster located directly adjacent to a stairwell. Due to the low AER, there is strong spatial heterogeneity in concentrations. The air flow inside the kitchen is buoyancy driven whereas convection driven flow is weak (Lai and Ho, 2008). Therefore air (and particles) make very strong upward flows due to this buoyancy force. The direct pathway for the particulate plume provided by the location of the toaster, and buoyancy driven flow created as a result of the toasting activity, generates rapid plume rise which is detected first by the upstairs (far-field) monitor before the plume evolves and is dispersed more widely around the kitchen and being detected by the near-field monitor. The open plan nature of House 1 could also in part account for the enhanced decay rate. The initial decay rate includes mixing of the air throughout the rest of the living space (larger mixing volume) so this could have increased overall ventilation rate. House 2, in contrast, and like most of the other houses studied, has a separate kitchen that can be isolated from the rest of the house, so the particles have longer to travel to reach the upstairs (far-field) monitor and since the pollution plume is not able to travel directly, dispersion and other removal processes occur first. Practically speaking, these results may be of particular interest to owners or occupiers of refurbished buildings, particularly where internal spaces have been reconfigured to accommodate multiple occupants, as well as provide guidance of new building design and planning decisions with respect to the placement and type of ventilation and appliances. Location of ventilation in relation to source around a residence is important (hence why we see longer decay rates upstairs or distal locations) as well as type, efficiency (CE) and venting ability amongst other factors.

4.5. Implications

We analysed trends in time-integrated measurements of cooking-generated PNC and manipulated the data in SPSS to examine differences per metric between near and far-field monitors in a single room and in two rooms per house to differentiate between individual houses and between individual scenarios within a single house.

Results highlight that residents can be exposed to high levels of UFP, up to 100 times background concentrations when cooking in a poorly ventilated kitchen. These UFP concentrations can persist for many hours after a cooking event in the absence of adequate ventilation, and extend beyond the kitchen, particularly once internal doors are opened, or in the case of an open-plan living space. Where the kitchen is not isolated from the rest of the house, UFPs generated by cooking activities can be transported to other rooms in the same house. Therefore, the health risk from cooking emissions may be underestimated if human exposure is only considered in the kitchen. Even though PNC were generally lower elsewhere in the house than in the kitchen, they can still be 100 times higher than background levels. Time spent in other rooms, such as upstairs (bedrooms) may be considerable (such as overnight) and therefore this is an important consideration.

UFPs are a serious health concern because of their small size and large surface area. The general public therefore needs to be reminded to ventilate effectively, either by natural or mechanical means, when cooking or undertaking other UFP-generating activities to avoid potentially detrimental health effects. Based on analysis of our data collected and our metrics, it appears that natural ventilation is generally more effective at removing particles than either no ventilation or mechanical ventilation for the majority of the time. Mechanical ventilation is also effective at reducing exposure to

cooking-generated particles, and we note that this strategy could be highly effective during the heating season when outdoor temperatures are low (hence the energy consequences of natural ventilation could be high) or where houses are built to airtight specifications. Widespread use of mechanical ventilation will impact upon the energy demand of the housing stock since range hoods use energy to move air and as such decisions about the design and use of such fans should consider the additional costs of IAQ improvement (Rim et al., 2012). The trade-off between improved IAQ and increased energy use will be explored in the next chapter. We have learnt a lot about the effectiveness of ventilation across our 8 houses with their different characteristics – both in terms of overall efficiencies (natural, mechanical and no ventilation) and effectiveness in reducing PN in the near field and far field depending on the layout of the house.

In this study we highlight the importance of ventilation with regard to ultrafine particulate pollution generated by cooking activities. However, the importance of ventilation is not limited to such pollutants, and ventilation is indeed significant in indoor air improvements with regards to many viral, chemical and particulate pollutants generated by a range of natural and anthropogenic sources. Ventilation has taken on increased importance in the current coronavirus pandemic as a means of reducing the risk of transmission in indoor spaces.

4.6. Limitations

This pilot study only considered a limited number of properties due to time constraints and invasive nature of monitoring in real-world environments. This limited our ability to find statistically significant differences between houses based on size, age, and other characteristics. Indeed, some parameters need careful interpretation, for example, a

small house can contain a large kitchen whereas a large house can contain a small kitchen. Since the experiments were conducted under real-world conditions, there are many confounding and influencing factors that could have influenced pollutant concentrations, some of which could not be controlled, making interpretation difficult. It would therefore be beneficial to replicate the cooking and ventilation scenarios described here in a controlled environment. The following chapter describes how these scenarios were replicated in the Salford Energy House, a specialist test facility.

4.7. Conclusions and Recommendations

This pilot field campaign involved deployment of novel multiple high resolution air quality monitoring devices across multiple residences in the NW of England to examine the particle response to discrete cooking events and the influences on the temporal and spatial patterns of this response including mitigation responses (natural and mechanical ventilation) and housing layout.

During episodic cooking activities, large increases of PNC occur. Peaks in concentration were generated quickly after the onset of cooking and decayed at a rate mainly determined by the air exchange rate and to a lesser extent, deposition on interior surfaces. We use 5 key metrics to evaluate differences between different source types and different forms of ventilation: Time to Peak (TTP), Peak Concentration (PKC), TTB (Time to Background), RTE (Rate of Decay) and Area-Under-Curve (AUC). TTP, PKC and AUC provide interesting insights into source strength and source variability within a single room or across multiple rooms. RTE and TTB provide interesting insights into the impacts of ventilation and other removal processes. We conclude there are significant differences in particle response with regards to ventilation. UFP emissions arising from our discrete cooking events suggest that cooking in a house with

inadequate ventilation could lead to indoor concentrations that significantly exceed those outside and could negatively affect the health of occupants. A cooker hood can be effective in reducing ultrafine emissions into the kitchen environment and preventing UFPs persisting for extended periods of time. However, the most effective strategy to reduce exposure to ultrafine emissions is clearly natural ventilation.

When we independently measured PNC at two locations in a single room, we concluded that a single V2000 monitor could give us a representative measure of IAQ. The exception to this is houses with open plan layouts, where airflows can be very different. In multi-room scenarios where we deployed multiple V2000 monitors around a house, we found cooking-generated particles at high concentrations elsewhere around the house and conclude in most instances that metrics derived from our near- and far-field monitors are generally statistically significantly different from each other. Higher peak concentrations and shorter times to peak were experienced in the far-field (upstairs) location in House 1, contrasting results from all the other houses which we see as the norm. We explained that this was a consequence of a buoyancy dominated flow and favourable housing layout and geometry.

This study provides insights into controlling influences on cooking emissions and the influence of housing dynamics, though the limited sample size inhibits firm conclusions. We recommend that this research could be repeated in the future, adopting a similar protocol but within a greater number and range of houses to enable statistical analysis to assess and evaluate the influence of housing characteristics on pollutant concentrations. It would also be beneficial to have more information on particle mass and particle size distributions and how these change over the course of a cooking event to interpret particle dynamics and understand processes at play including deposition and

coagulation more quantitatively. This, however, comes with its own shortfalls and difficulties in accessing properties without significant disruption, which we did. Whilst natural ventilation is seen as the most effective strategy for reducing exposure to cooking-generated particles, we understand that this has energy implications, and we need to consider the quality of the ambient air. Owing to temperature and pressure differentials, ventilation heat loss from natural ventilation can be considerable, especially during cold winter periods, and the energy consequences of reheating lost air could be high. This will be the focus of research presented in the next chapter.

5.0. Exploring the Trade-Offs Between Good Indoor Air Quality and Energy Efficiency in a Specialised Test Facility

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Manuscript has been prepared for submission to Building and Environment. This study was conceived and designed by PhD Student Charlotte Farr with extensive collaboration from Salford University at the Salford Energy House. The air quality section of this manuscript was written by Charlotte Farr with editing and corrections made by supervisory team at Lancaster University; Duncan Whyatt, Andrew Sweetman and Douglas Booker. CO_2 decay measurements and energy penalties were calculated by David Farmer of Salford University, who also wrote sections of the methodology and contributed towards the results.

Abstract

Residential cooking activities generate significant amounts of particles that may cause serious health effects. Epidemiological studies highlight the importance of good exposure mitigation measures to reduce acute or chronic health effects. Natural and mechanical ventilation have been shown to be effective in reducing pollutants below harmful levels, however, both increase air exchange with the external environment, leading to heat loss, the extent to which varies depending on ventilation type and ventilation period. This pilot study replicates residential cooking activities within the Salford Energy House under different ventilation regimes to assess trade-offs between indoor air quality and energy efficiency. We focus on assessing exposure mitigation to cooking generated particles by calculating energy penalties associated with heat loss and determine which strategies and combinations of behavioural and technological

interventions can simultaneously balance the competing demands of reducing pollution levels whilst maintaining energy efficiency.

We find that natural ventilation results in modest energy penalties, which increase for longer periods of ventilation, and suggest that windows may be opened for up to 20 minutes to improve indoor air quality after cooking with no significant loss in energy. We indeed observe an energy surplus in some instances when considering the additional energy generated by cooking processes. We find that mechanical ventilation results in lower energy penalties than natural ventilation for all periods. We suggest an optimum of 20 minutes of extract operation, which is when we observe the highest particle decay rates, which yields good IAQ benefits at little detriment to energy efficiency. This pilot study makes key associations between indoor air quality and energy efficiency which has wider application. The findings and implications are only applicable to houses similar to the test house under certain environmental conditions.

Keywords: indoor air quality, ultrafine particles, ventilation, heat loss, energy penalties

5.1. Introduction

The domestic sector is responsible for 32% of UK energy consumption, most of which is attributable to space heating demand (BEIS, 2018). As such, public policies and programs have been introduced to address decarbonisation through promoting energy efficient buildings (Persily and Emmerich, 2012), including the EC Energy Performance Building Directive (EPBD, 2010/31/EU). Energy saving measures within residences target improvements in the thermal integrity of the envelope, as well as increasing the efficiency of heating and cooling equipment and reducing system energy use (Persily and Emmerich, 2012; Vasile et al., 2016). Recent legislation focuses on

increasing insulation levels and improving airtightness to prevent uncontrolled heating losses (Shrubsole et al., 2014). Whilst building standards have been established, many residential buildings, especially older ones, do not meet these standards (Fortenberry et al., 2019). Problems with the thermal integrity of the envelope cause high infiltration losses and low energy efficiency in the existing building stock (Fitton et al., 2014). Therefore, to meet energy efficiency targets the energy performance of nearly all dwellings needs be improved by 2030 (Hamilton et al., 2017).

Ventilation, which involves introducing and circulating fresh air through a building and removing or diluting contaminated air is important for a healthy building (Dimitroulopoulou, 2012). However, due to the act of ventilation and impacts on the heat balance, ventilation makes up a large proportion of the energy consumption in buildings and as such is an attractive target for energy saving (Guyot et al., 2018). Traditionally, natural ventilation, driven by wind and thermally generated pressures, has met ventilation needs (Dimitroulopoulou, 2012). However, natural ventilation is difficult to control due to reliance on unreliable driving forces, which can result in periods of insufficient ventilation followed by periods of over-ventilation and excessive energy waste (Liddament, 1996). In contrast, mechanical ventilation systems provide a controlled rate of air change in response to the varying occupant needs and pollutant loads (Liddament, 1996).

Mechanical ventilation, airflow in and out of a building caused by a fan through intake and/or exhaust vents (Seppänen, 2008), adds to the energy demands of a building. In countries with colder climates, where houses need to be airtight to conserve heat, whole house mechanical ventilation systems are more prevalent. In Britain's temperate climate, houses used to be so leaky that whole house mechanical ventilation was not necessary.

However, as new builds are more airtight, these systems are now commonplace. Policies that focus on air tightness, while decreasing energy requirements have a tendency to decrease ventilation rates at the possible expense of adequate indoor air quality (IAQ) (Sherman and Matson, 1997). Improving building airtightness without providing additional ventilation can lead to lower ventilation rates and poorer IAQ (O'Leary et al., 2019b).

Studies exploring the relationships between energy efficiency and air quality are on the rise. The conflict between a desire to minimise ventilation rate to reduce energy demand, and to maximise ventilation rate to improve IAQ remains a pressing issue (Liddament, 1996). Increasing the ventilation rate either naturally or mechanically will reduce indoor-generated pollutant concentrations but will result in energy 'penalties' (Dimitroulopoulou, 2012). Epidemiological effects have been studied, with mixed results. Low ventilation rates (at or below 10 l/s) may elevate concentrations of indoor-generated pollutants which are associated with sick building syndrome and detrimental health effects (including inflammation, infections, asthma, allergy) (Seppänen et al., 1999; Dimitroulopoulou, 2012).

Health gains in Europe attributed to effective implementation of the Energy Performance Building Directive, which includes IAQ issues, have been estimated at more than 300,000 disability-adjusted life years per year (Guyot et al., 2018). Ventilation should not be seen as a complete solution, however; to achieve good IAQ, source control must also be considered (Guyot et al., 2018). Eliminating sources or using low pollution products and materials where possible could lower ventilation energy requirements (Seppänen, 2008). However, it is not always feasible to eliminate pollutant generating activities completely.

Cooking generates a significant amount of organic and non-organic gaseous and particle pollution and is regarded as a major source of pollutants in residential environments (Lai and Ho, 2008; Laverge et al., 2011). These pollutants can reach hazardous concentrations in the kitchen space as well as elsewhere throughout a house (Logue and Singer, 2014). Particles generated by combustion related activities such as cooking are generally within the ultrafine (UFP thereafter; diameter < 0.1 μm) and fine ($PM_{2.5}$; diameter < 2.5 μm) particle size ranges (Abdullahi et al., 2013). Due to their small size, these particles exert higher toxicity than larger particles, as epidemiological literature demonstrates (Ohlwein et al., 2019). This is concerning as peak particle number concentrations (PNC) associated with cooking in the indoor environment have been found to be higher than peak concentrations in the outdoor environment (Dennekamp et al., 2001).

Researchers have investigated cooking emissions and influencing factors. Studies have been carried out in a) real-life kitchens, where the emissions are influenced by many factors including room arrangement, building materials, outdoor infiltration, other combustion devices, ventilation and cooking methods, and in b) controlled chambers, where there are fewer external influences and emissions are influenced by the type of fuel and the type of food being cooked (Hubuyo et al., 2011). Reported emission rates for the cooking of single ingredients (Dennekamp et al., 2001; Isaxon et al., 2015; O'Leary et al., 2019a; Afshari et al., 2005; Wallace et al., 2011) and full meals (He et al., 2005) are highly variable.

Simply reducing or eliminating the processes involved in cooking in order to improve IAQ is unrealistic and undesirable. Cooking is conducted on an almost daily basis in most residences (O'Leary et al., 2019a) and is necessary for the safety and enjoyment

of a substantial number of food products (Hager and Morawicki, 2013). Therefore, mitigation measures need be considered, most importantly the use of a cooker hood (mechanical extract ventilation), and secondarily the use of non-stick frying pans and cooking methods that avoid the browning or charring of food (O'Leary et al., 2019a). Natural ventilation is also effective; however, this causes ventilation-related heat loss, particularly in the heating season**.** In the UK, kitchens in new dwellings have to include cooker hoods with intermittent extract rate of 30 l/s or have a ventilation rate of 60 l/s (UK Government, 2014a). However, there is no requirement to modify ventilation in existing dwellings (O'Leary et al., 2019a).

Range hoods mounted over the cooktop should be used during cooking to maintain good IAQ by extracting pollutants at their source, as shown by both experimental and modelling studies (Logue and Singer, 2014; Dobbin et al., 2018). Venting range hoods that exhaust kitchen contaminants directly to the external environment (Kim et al., 2018), and that avoid pollutants mixing with kitchen air are most effective (Logue and Singer, 2014). Using extract ventilation during cooking is especially important in airtight dwellings and in other houses during the heating season when occupants seek to reduce ventilation rates to obtain thermal comfort and to minimise fuel heating costs (O'Leary et al., 2019a). Increasing range hood use will impact the energy demand of the housing stock, since range hoods use energy to move air (Logue and Singer, 2014). Further work is required to estimate how mechanical ventilation devices would affect the housing stock energy demand in the UK (O'Leary et al., 2019b).

There has been a lack of research evaluating exposure mitigation to UFP due to the invasive nature of such studies in real-world environments. This pilot study replicates a series of short-duration cooking activities in a specialist test facility to assess the

effectiveness of different forms of ventilation for exposure mitigation and examine the trade-offs between good IAQ and energy efficiency. This is the first study of this kind to be conducted in such an environment. The test facility provided us with a unique environment in which to measure PNC, space heating and ventilation heat loss in unprecedented detail to gain an understanding of the energy penalties associated with different mitigation strategies**.** Our objectives were as follows:

1. Use high resolution air quality monitors to monitor IAQ in a specialised test facility;
2. Examine PNC response to discrete cooking activities under different ventilation scenarios;
3. Examine the energy penalties of exposure mitigation through measurements of spacing heating and ventilation heat loss;
4. Explore IAQ-energy efficiency trade-offs with time from the onset of cooking.

5.2. Materials and Methods

5.2.1. Salford Energy House

All testing for this study was based at the Salford Energy House. Built in 2011, the Energy House is a full-sized two-bedroom end of terrace house, typical of Salford in 1919 (Fitton et al., 2014). It was built using reclaimed materials inside a climate-controlled chamber which can replicate a wide range of weather conditions (Figure 5.1). In this study the chamber was set to 5.6 °C, typical of wintertime in the UK, when the trade-offs between reducing cooking related air pollution exposures and ventilation related heat loss are potentially at their greatest. Internal and external conditions were kept the same for the IAQ and ventilation rate measurements which were conducted on different dates. The Energy House has solid brick walls, suspended timber floors, lath

and plaster ceilings, single glazed windows and is uninsulated. Heating is provided by a wet central heating system. The house is classed as 'hard-to-treat' in terms of energy efficiency due to the lack of cavity walls. Therefore, it experiences higher rates of air infiltration and has a lower energy efficiency rating than newer housing (Fitton et al., 2014). It is fully furnished and numerous sensors in each room monitor a range of variables (including temperature, air flow velocity, boiler power and energy usage) using a custom time series program to provide real time analysis (Fitton et al., 2014). Further details of this test facility are discussed in Fitton et al. (2014).

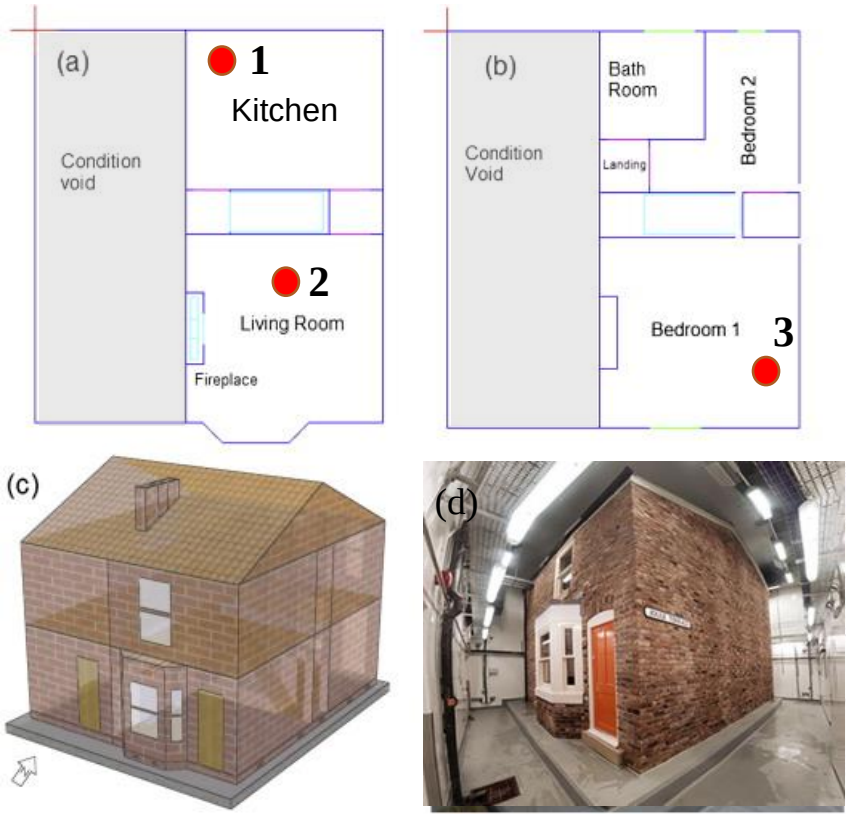


Figure 5.1: The Energy House ground (a) and first (b) floor plans, (c) the 3D Energy House model and (d) the Energy House in the chamber (Fitton et al., 2014). Locations of air quality monitors are shown in red. Conditioning void attached to the house, is used to replicate typical end-terrace environment.

5.2.2. Air Quality Monitoring

This study utilised NAQTS V2000 air quality (V2000 thereafter) monitoring units. These units house a variety of air quality and environmental sensor which can be used to capture data at high (1-second) temporal resolution. Particle number concentrations (PNC) were monitored with a condensation particle counter (CPC) housed inside the V2000 unit. The CPC does not have a size selective inlet hence counts particles below 2.5 µm, however most freshly created particles from cooking are indeed UFP. It is also well known that combustion-generated particles are considerably smaller than 2.5 µm, which justifies monitoring PNC of UFP to determine occupant exposure to cooking-generated particles (Isaxon et al., 2015).

During monitoring in the Energy House, the V2000 units were placed in different rooms as illustrated in Figure 5.1. Initially the units were run to establish the background PNC. They were then used to capture PNC for 3 sets of experiments (Table 5.1). Initial experiments were conducted in the kitchen (volume of 35 m^3) with the internal door to the living room closed. Firstly, bread was toasted for 5 minutes on the highest setting under different ventilation scenarios i.e., no ventilation, then natural ventilation with the kitchen window opened to its full extent for different periods of time (5, 10, 20 and 40 minutes). Natural ventilation was used once cooking had ceased. Secondly, a single egg was fried over an electric hob for the same ventilation scenarios plus a mechanical extract ventilation scenario. Mechanical extract ventilation was used from the onset of cooking for 30 minutes. Finally, the toasting experiments were repeated, but with internal doors in the Energy House opened to promote free movement of particles elsewhere around the house. This increased the volume for dispersion and changed airflow throughout the house and subsequently opportunities for ventilation.

Table 5.1: Replicated episodic cooking activities (toasting and frying) over the four consecutive monitoring days at the Salford Energy House. Further experimental details; number of monitors, ventilation scenario and period, and housing configuration are given.

Experiment	Cooking	No. Monitors	Ventilation	Ventilation Period	Door Status
1A	Toast	1	None	-	Closed
1B	Toast	1	Natural	Full Duration	Closed
1C	Toast	1	Natural	20 Minutes	Closed
1D	Toast	1	Natural	10 Minutes	Closed
1E	Toast	1	Natural	5 Minutes	Closed
2A	Frying	1	None	-	Closed
2B	Frying	1	Natural	Full Duration	Closed
2C	Frying	1	Natural	20 Minutes	Closed
2D	Frying	1	Natural	10 Minutes	Closed
2E	Frying	1	Natural	5 Minutes	Closed
2F	Frying	1	Mechanical	-	Closed
3A	Toast	3	None	Full Duration	Open
3B	Toast	3	Natural	20 Minutes	Open
3C	Toast	3	Natural	5 Minutes	Open

Before each test, the pans and cooking utensils were cleaned in warm water and hand dried. At the end of each cooking period, all burners were turned off and a lid was placed on any frying pan to prevent continued emissions, similar to O'Leary et al. (2019a).

5.2.3. Statistical Analysis

PNC over time were visually assessed during each cooking experiment. Descriptive statistics similar to those used in hydrology and in the previous chapter of this thesis, enabled comparisons of metrics of time to peak, peak concentration and time to background between ventilation scenarios. A decay rate was estimated for every experiment where a clear rise and fall in PNC could be observed by fitting a linear regression of the natural logarithm (Dobbin et al., 2018). The impacts of any outliers were eliminated by taking temporal measurements 5% below the peak and 5% above background. Extending over longer time scales can result in deviations from linearity

(Dobbin et al., 2018). UFP source strength and emission rates were calculated similar to O'Leary et al. (2019a). The area-under-the-curve of a plot of concentration over time is equivalent to the total number of particles emitted, i.e., a surrogate of the source strength. An emission rate was calculated as the source strength divided by the time (duration) of cooking activity.

5.2.4. Energy Penalty Measurements

For the purpose of this study, an energy penalty is defined as the additional energy consumption resulting from actions undertaken to reduce the concentration of cooking generated pollutants within a dwelling by means of increasing the rate of ventilation with the external environment. This study considers increases in energy consumption attributable to space heating demand and the operation of natural and mechanical ventilation systems. We calculate energy penalties that both exclude and include additional energy inputs from the cooking activities themselves. The energy penalty resulting from the ventilation regimes employed during the IAQ study was estimated using two methods: (a) measuring the increase in space heating energy consumption during a period of ventilation, (b) calculating the energy required to heat additional air exchange with the external environment based on ventilation rate measurements. These measurements were undertaken simultaneously after the IAQ measurement campaign and did not involve cooking.

The internal environment of the Energy House and its adjoining neighbour (conditioning void) were maintained at the temperatures used by the UK Government's Standard Assessment Procedure (SAP) methodology for calculating domestic energy use of 21 °C in the living room and 18 °C in all other zones (BRE, 2012). The chamber

temperature was maintained at 5.6 °C which can be considered representative of the external temperature during the heating season.

5.2.4.1. Space Heating Energy Measurements

At steady state, any change made to the ventilation of the Energy House should result in a change in space heating energy consumption in order to restore the steady state. In this case, opening the kitchen window should result in an increase in space heating energy consumption, the so-called 'energy penalty'. Conditioning of the internal and external environments commenced 48 hours prior to the ventilation measurements so that heat transfer between the internal environment of the Energy House and the chamber could be considered akin to steady state (Farmer et al., 2017).

Electric resistance heaters with thermostatic controllers were placed at the centre of each room to provide highly responsive temperature control and an accurate means of measuring space heating energy consumption, thus removing uncertainty regarding the sensitivity of thermostatic radiator valves and efficiency of the central heating system. Electricity consumption to the Energy House was measured at 1 Wh resolution at intervals of one minute using energy meters (uncertainty ± 1%).

5.2.5. Energy Penalty Calculations

Ventilation Rate Measurements

This part of the study was designed to measure the increase in the ventilation rate resulting from each intervention. The energy penalty can then be obtained by calculating the energy required to heat the additional air exchanged with the external environment.

Natural Ventilation

The energy penalty for a window opening ventilation period was obtained using the following equation;

$$E_p = (E_h - E_c) + E_v$$

Where:

E_p= Energy penalty that includes additional heat input from cooking appliance

E_h = Energy required to maintain kitchen air temperature with window closed (obtained from energy measurements (Section 4.2.4) with Energy House at steady state

E_c = Heating energy input from cooking device during cooking period

E_v = Energy required to heat additional air infiltration required for cooking ventilation (obtained from ventilation rate measurements)

E_v is calculated as;

$$E_v = \frac{\Delta n \, c_p \, \rho \, V \, \Delta T \, t_v}{3600 * 1000}$$

Where:

Δn is the measured increase in the air change rate within the ventilated space[1] (ACH)

c_p is the specific heat of air[2] (J/kg K)

ρ is the density of air[3] (kg/m^3)

V is the volume of the ventilated space[4] (m^3)

ΔT is the temperature difference between the set-point temperature and external environment (K)

t_v is the ventilation period (hours)

Δn is the measured difference of the air change rate (n) with the kitchen window in the open (cooking ventilation) and closed (baseline) positions. n was measured using CO_2 concentration decay measurements following the guidelines detailed in ASTM E741-11 (2017). The ventilation periods associated with the cooking experiments (Table 5.1)

[1] The 3600 value in the denominator accounts for the conversion of ACH to ACS and the 1000 value converts Wh to kWh.
[2] Specific heat of air at 5°C is 1002 J/kg K (Engineering Toolbox, 2020)
[3] Density of air at 5°C is 1.268 kg/m^3 (Engineering Toolbox, 2020)
[4] Volume of kitchen is 26.92 m^3

were deemed to be too short to obtain confident measurements of n. Therefore, a ventilation period of one hour in duration was chosen to undertake the CO_2 concentration decay measurements. This is considered an acceptable approach for deriving n for shorter periods as the n should not vary with time due to the presence of constant conditions within the environmental chamber. The measurements were undertaken during the space heating energy monitoring period. The procedure for the CO_2 concentration decay measurements was as follows. A Sauermann Si-AQ Expert IAQ monitor (uncertainty ± 2%), set to record CO_2 concentration at intervals of 30 seconds, was positioned in the centre of the kitchen. A fire extinguisher was used to release a burst of CO_2 into the kitchen. A two-minute period followed in which an air circulation fan was used to mix the air within the kitchen. The Energy House was then vacated, and the kitchen window opened using the building control system for a period of one hour (the window remained closed for the baseline measurements). n was calculated using the method detailed in the equation below.

$$n = \frac{\ln C(t_2) - \ln C(t_1)}{t_2 - t_1}$$

Where:

n is the air change rate (ACH)

C is the CO_2 concentration measurement

t_1 is the start of the CO_2 concentration decay analysis period

t_2 is the end of the CO_2 concentration decay analysis period

A linear regression analysis of the log of the normalised CO_2 concentration (C_n) against time was also performed to test the assumption than n remained constant throughout each measurement period. C_n was calculated using equation (Roulet and Foradini, 2002);

$$C_n = \frac{C(t) - C(t_1)}{C(t_1) - C_b}$$

Where:

C_b is the background concentration level

t is the time of measurement

Mechanical Ventilation

The energy penalty for a mechanical extract ventilation period was obtained using the following equation;

$$E_p = (E_h - E_c) + E_v + E_f$$

Where:

E_v = Energy required to heat additional air infiltration required for cooking ventilation (obtained grille flow measurement of extractor fan)

E_f = Energy required for mechanical ventilation over operational period (based on manufacturers literature value of 22.6 W)

5.3. Results and Interpretation

In this section we first present the results of our PNC measurements under different ventilation scenarios, then the results of our energy penalty calculations before going on to synthesise trade-offs between IAQ and energy efficiency in the discussion.

5.3.1. PNC response to ventilation within the kitchen

All the PNC profiles show similar trends (Figure 5.2). As time progresses after the onset of cooking (whether toasting or frying) the concentration remains low for a brief period of time, then rises sharply, observed as a rising limb. PNC increases with time as cooking continues, indicating ongoing emissions of UFP until a peak concentration is

reached. Once cooking has stopped, the PNC decays exponentially towards background concentrations with the rate largely governed by air exchange rate.

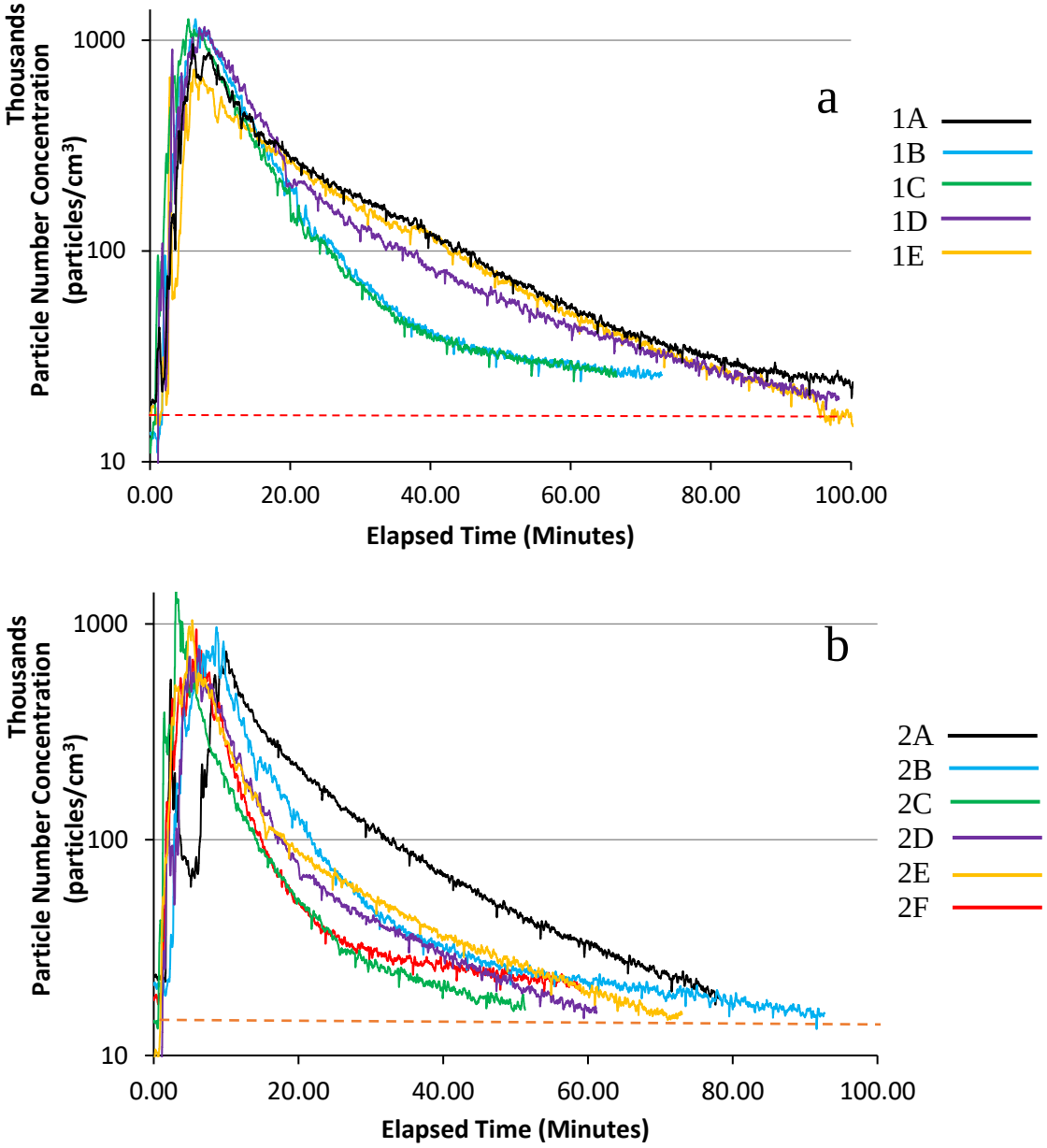


Figure 5.2: Temporal particle number concentration (PNC) resulting from episodic cooking experiments under different ventilation scenarios for (a) toasting and (b) frying in the EH. Refer to Table 5.1 for details of individual experiments and ventilation scenarios corresponding to each scenario.

PNC profiles over time highlight significant differences between ventilation scenarios (Figure 5.2). The concentration profile is a product of many factors including the cooking method, particle size, the relative location between the source and the V2000

monitor and the indoor airflow (Lai and Ho, 2008). In this study, since the cooking methods are constant, ventilation rate is the dominant force controlling indoor airflow, thereby time to peak. We observe little difference in time to peak between ventilation scenarios. Time to peak for toasting experiments was ~5 minutes for each ventilation scenario, highlighting fast air mixing. Time to peak was slightly longer for egg frying experiments, reflecting the longer event duration. Nonetheless it was fast at around 10 minutes, expected due to the high oil temperature and large heated surface.

Each cooking experiment increased PNC to at least a hundred times background levels. PNC remained at this magnitude for approximately 5 minutes for toasting and slightly longer for frying, reflecting the longer duration of this event, before declining. Peak PNC differed between experiments, despite each being conducted in the same way, reflecting the influence of ventilation (and perhaps some variations in source strength). Decay to background concentrations took up to 100 minutes.

Our study aimed to investigate different rates of air exchange caused by different ventilation regimes and the consequential effects on decay rate. The background air exchange rate, estimated from the CO_2 measurements reported in the previous section was 2.3 air changes per hour (ACH), indicating the Energy House is “leaky” compared to newer housing stock and there may be higher infiltration losses. However, the main variable to change between tests was the air exchange rate provided by natural or mechanical ventilation. Air exchange rates were higher when windows were opened (7.4 ACH) or the extract ventilation was operational (7.0 ACH). Enhanced ventilation provided by natural ventilation or mechanical ventilation ensured that peak concentrations were not maintained for long periods of time. Higher ACH provided by ventilation promote indoor-to-outdoor transport and faster dispersion of pollution loads.

The time taken for PNC to decline from peak to background levels influences a person's potential exposure to cooking generated particles. The longest time to background is associated with the non-ventilated scenarios (Figure 5.3), taking 80 minutes for frying and 100 minutes for toasting, in direct response to lower air exchange rates, because the indoor airflow is lower, and air exchanges between the kitchen and the chamber is solely dependent on infiltration. Due to the property age and background ACH, larger infiltration losses could be expected, promoting faster decay but this does not appear to be the case. For toasting, much shorter times to background for naturally ventilated Scenarios 1B and 1C are observed than Scenario 1A showing that window opening is highly effective at removing particles from the kitchen environment and reducing potential exposures.

We see ventilation as a significant influence on decay rate, and the influence of deposition is much less significant. This is indicated by the differential decay rates between non-ventilated scenarios (where removal mechanism is mainly deposition) and ventilated scenarios (where removal mechanisms comprise dispersion across space and deposition) and assertions we made in the previous chapter (Section 4.3.1). Particles smaller than 2.5 μm are largely uninfluenced by gravitational forces and have very low settling velocities (Liddament, 1996). They tend to remain in suspension by continual molecular bombardment and display diffusion properties similar to a gas (Liddament, 1996). Since the majority of freshly generated particles by cooking are smaller than 1 μm therefore we expect insignificant gravitational settling.

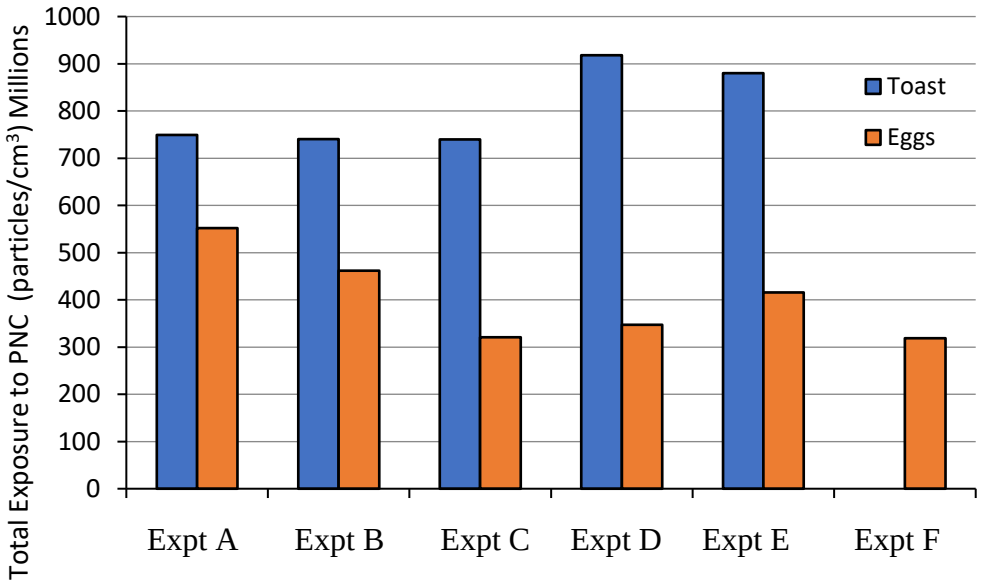
The rate of particle decay from peak to background concentration highlights slower rates of decay for the non-ventilated scenarios because of the reduced airflow. The decay rate is fairly uniform for most scenarios especially those non-ventilated, however

for some naturally ventilated scenarios; 1B, 1C and 1D in particular the decay appears rapid at first but is then slower as PNC decline to background levels. In essence we are seeing a much higher initial decay rate (first 20 minutes) relative to subsequent and overall average decay period (Appendix B). The initial decay rate is likely to be dominated by dispersion, whilst other processes, such as coagulation and deposition are more likely to explain the subsequent slower decline to background levels. It was expected that opening the window for the longest period would elicit the fastest decay rate due to greater ACH between the kitchen and the chamber. This was true for toasting, but not for frying, with Scenario 2B (windows open for full duration of cooking experiment) producing a slower decay rate than experiments with shorter ventilation periods. We find that for frying based on assessment of decay rates and temporal PNC data mechanical ventilation is the most effective mitigation strategy. This is particularly evident for the first 20 minutes (initial decay rate) where we observe even further enhanced decay. Based on decay rates we conclude scenarios 1B and 2F are most effective after toasting and during frying respectively for improving IAQ and reducing potential exposures to UFP.

Experiments 1B and 2B and 1C and 2C show the greatest range of PNC and the highest peak concentrations. This is unexpected since higher ventilation rates would be thought to promote greater air exchanges and suppress high peak concentrations after cooking (Appendix B). Experiments 1A and 2A (no ventilation) show the smallest range in PNC and the lowest peak concentrations, again unexpected since under these scenarios it would be expected that low air exchange rates would promote elevated concentrations. These anomalies require further investigation but could perhaps reflect higher rates of deposition in the absence of higher airflow and coagulation, as well as infiltration through the building envelope. They could also reflect indoor airflow changes between

various internal environments, with and without the presence of air exchange with the external environment.

The plot of concentration over time is equivalent to the total number of particles emitted, and is taken as a representative measure of exposure and surrogate for source strength (Figure 5.3). The emission rate was calculated by dividing the UFP source strength (area-under-the-curve) by the emission period (O'Leary et al., 2019a). UFP source strengths are similar for each toasting event, which is expected since each experiment was conducted in the same environment in the same manner. The source strengths and emission rates showed more variability for the frying experiments. Despite the shorter duration of the experiment, toasting generated more particles than frying in every instance.



Toasting Emission Rates (cm^3/s)	2,500,000	2,500,000	2,500,000	3,000,000	3,000,000	
Egg Frying Emission Rates (cm^3/s)	1,200,000	1,000,000	700,000	700,000	900,000	700,000

Figure 5.3: Exposure characteristics: area-under-the-curve quantification and emission rates for each ventilation scenario (A–F) for each cooking experiment (toasting and frying). Emission rates are rounded up to the nearest hundred thousand.

5.3.2. PNC response to ventilation across whole house

The PNC data from one near-field (kitchen) and two far-field (living room and upstairs) V2000 monitors can tell us something about how cooking activities can impact PNC around a house and the relationship between ventilation and airflow throughout the house more generally (Figure 5.4). We see that we significantly underestimate the effect of cooking-generated particles if we do not consider pollution levels in other rooms of the Energy House. Though much lower than in the kitchen, high PNC in the living room suggest passive transfer of pollutants and larger than expected indoor airflow velocities despite internal doors being closed. This may be a cause for concern due to their persistence. Higher PNC have been found in houses with open plan kitchens (Nasir and Colbeck, 2013) so it is not surprising to observe even higher concentrations than previously found throughout the house when the interior doors are opened. Peak PNC differs significantly between the near and far-field locations. For the ventilated scenario, the cooking peak is highest in the kitchen, lower in the living room and lower still in the bedroom, as we expect and was similarly observed by Dimitroulopoulou (2012). In general, the longer the particles travel, the greater the impact of removal processes such as deposition and coagulation. For the non-ventilated scenario, it is the upstairs (far-field) location that experiences the highest peak concentration, suggesting that in the absence of air exchange between the kitchen window and external chamber there are higher indoor airflows. This is interesting because it makes us also consider the importance of air exchange between rooms as much as between internal and external environments.

Higher decay rates throughout the house indicate higher indoor airflows when the interior doors are opened (Appendix B). Part of the initial decay rate includes mixing

of the air throughout the rest of the living space (larger area for dilution), so faster rates of decay are expected, particularly in the kitchen. Since ventilation is only experienced in the near-field, the ventilation scenario is less of an influence on PNC in other rooms of the house.

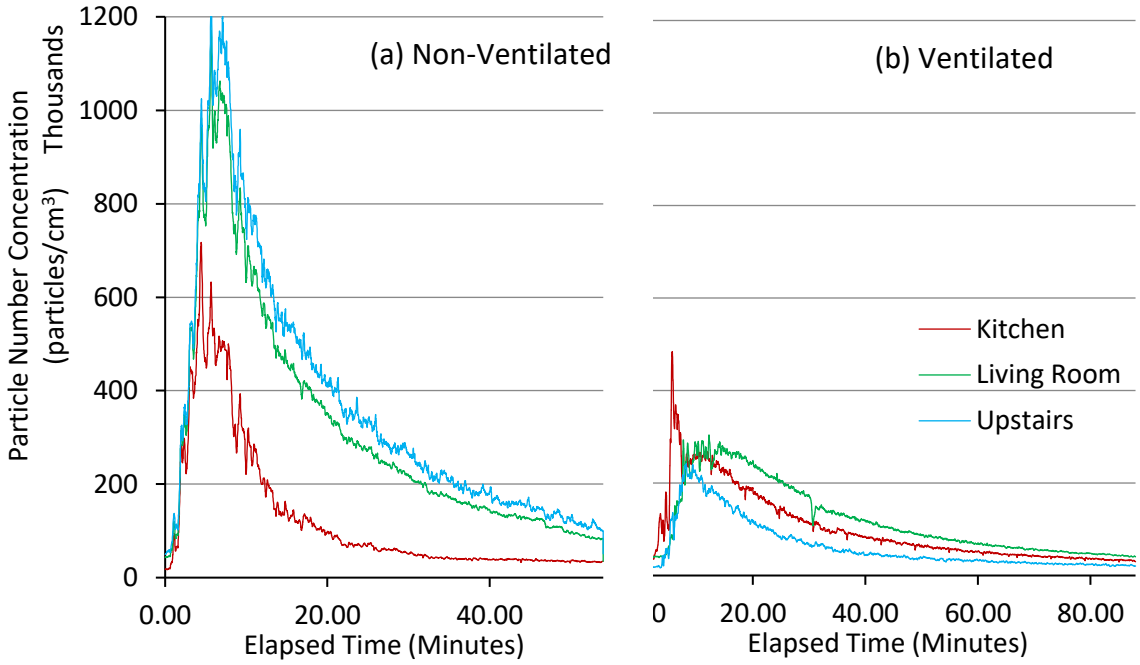


Figure 5.4: Temporal trends in PNC at both near field (kitchen) and far-field (living room and upstairs) locations resulting from toasting under non-ventilated (Expt 3A) (a) and ventilated (Expt 3B) (b) scenarios with internal doors within the Energy House open.

5.3.3. Energy Penalties

To reiterate, an energy penalty is defined as additional energy consumption resulting from actions undertaken to reduce the concentration of cooking generated pollutants within a dwelling by means of increasing the rate of ventilation with the external environment. We calculated energy penalties using data derived using 2 different methods.

Method 1: Space Heating Consumption

Figure 5.5 shows the space heating energy consumption for a) the kitchen and b) the entire Energy House during each hour after the 48-hour steady-state stabilisation period including two one-hour periods of window opening. Table 5.2 provides the mean space heating energy consumption for all periods with the kitchen window in both closed and open positions (excluding the hour following ventilation).

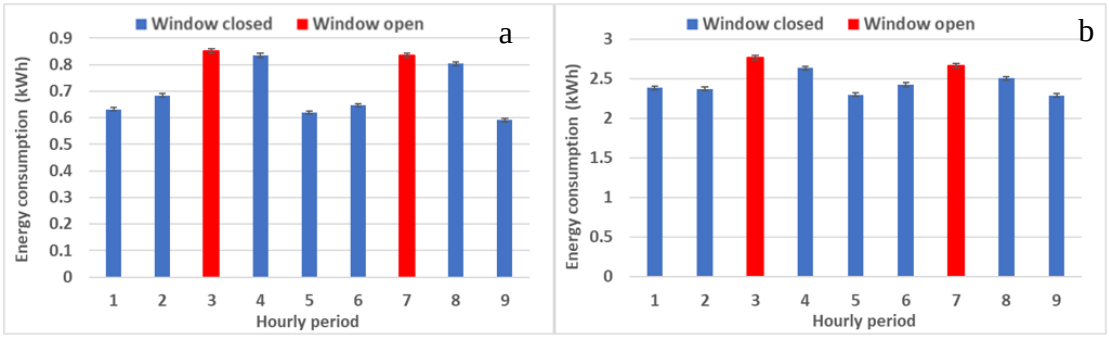


Figure 5.5: Hourly space heating energy consumption for (a) the kitchen and (b) the entire Energy House during hourly periods with the kitchen window open and closed.

Table 5.2: Mean space heating energy consumption for the kitchen and the entire Energy House for periods with the kitchen window in both the closed and open position.

Window position	Space heating energy consumption (kWh)	
	Kitchen	Entire Energy House
Closed	0.634 (± 0.006)	2.452 (± 0.025)
Open	0.843 (± 0.006)	2.663 (± 0.027)
Difference	0.209 (± 0.008)	0.211 (± 0.037)

It can be seen in Figure 5.5 that the two periods where the kitchen window was open both coincided with elevated space heating energy consumption in the kitchen and throughout the entire Energy House. This elevated energy consumption during window opening is in response to ventilation related heat loss. The space heating energy consumption measurements show an average increase in space heating energy consumption of 0.21 kWh during a one-hour period of window opening, all of which can be attributed to additional kitchen space heating consumption. The increase in

energy consumption is entirely attributable to the electric heater during and after the window was opened. The increase is significant but lower than that derived from ventilation measurements (see results of Method 2).

Figure 5.5 also shows that kitchen space heating energy consumption remained elevated during the one-hour period following window opening. We assume this is because of adjustment and continued indoor airflows within the house. After the window was closed the temperature in the floor void remained lower for a period and this required additional heat input. Temperature measurements suggest that opening the kitchen window increased air flow through the entire Energy House, some of which was through the underfloor void into the kitchen. The reason for this behaviour could be due to cooling of the suspended timber ground floor during the window opening period: air temperature measurements in the suspended timber ground floor void below the centre of the living room during window opening periods recorded a 0.1 °C reduction from ~ 10 minutes after the window opening until ~15 minutes after the window closed. It is likely that the air temperature in the kitchen floor void (not measured at the time of study) experienced a greater reduction. The air temperature of the loft also increased by 0.1 °C ~30 minutes after window opening until ~15 minutes after closing. This suggests that window opening not only resulted in additional ventilation through the window, but also increased the drivers for ventilation throughout the dwelling (through enhancing the stack effect, caused by thermal differences). The energy and temperature measurements suggest that air from the chamber was also drawn into the underfloor void via airbricks within the void walls, into the kitchen through gaps in the fabric, then passed around gaps around the door and other paths within the building fabric to the upper levels of the Energy House. This observation provides an explanation for the

higher concentration of cooking-based pollutants measured by the upstairs monitor with the kitchen window open during IAQ measurements (Figure 5.4).

We conclude that the one-hour period used to measure an energy penalty from space heating energy measurements was insufficient in duration to allow the EH to reach steady-state again or to account for the complexity of air exchange paths between the internal and external environment that resulted increased space heating energy consumption after the window was closed. This means that any energy penalty derived from this method is likely to be an underestimation as it fails to account for dynamic processes. For this reason, we now go on to use the ventilation rate measurements, which should yield more robust results.

Method 2: Ventilation Rate Measurements

Window Opening

In this instance we use ventilation rate measurements (defined in the methodology) to estimate ventilation heat loss and energy required to heat incoming air for window opening periods (Figure 5.6). The measurements show that there was a very close agreement between n and C_n (Table 5.3). This suggests that the ACH during each scenario remained constant and that it is acceptable to base energy penalties for periods < 1 hour in duration on ACH measured over a one-hour period. The value for n with the window in the closed position is in good agreement with the assumed value for kitchens within housing of this age group contained within the 2017 CIBSE Domestic Heating Design Guide of 2 ACH.

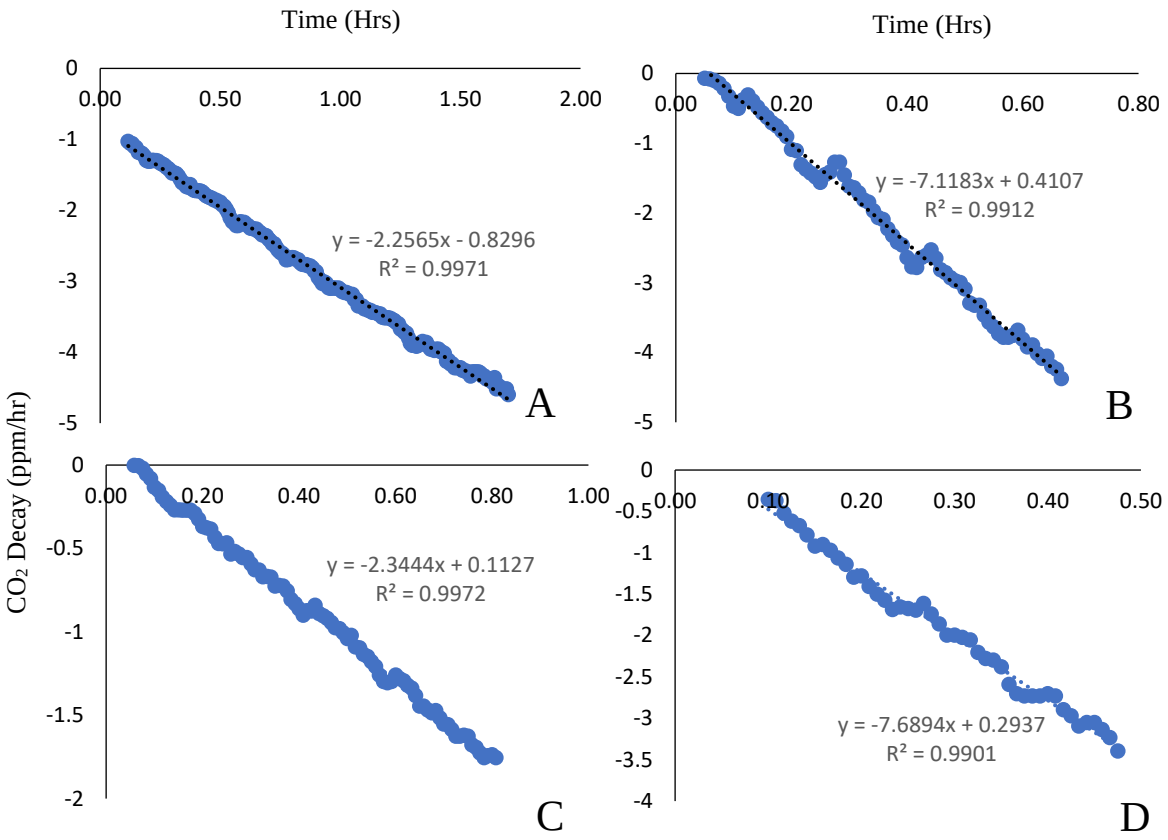


Figure 5.6: CO_2 decay rates used to calculate ACH when windows were closed (**A** and **C**) and when windows were open (**B** and **D**).

The increase in energy consumption derived from ventilation rate measurements for a one-hour period of window opening is three times greater than the increase in space heating energy consumption measured during a one-hour window opening period that we identified previously.

If the one-hour period following window opening (during which space heating energy consumption was elevated) is also included in the space heating energy measurement, then energy consumption for a one-hour period derived from the ventilation rate measurements is still 1.5 times greater reflecting uncertainties around the validity of the space heating energy consumption approach and how this method failed to account for time lags within the system. We therefore decided to use ventilation rate measurements to estimate energy penalties for natural and mechanical ventilation, since the space heating consumption methodology appeared to underestimate these penalties.

In Table 5.3 we calculate the energy required to heat the incoming air for different periods of window opening, without considering additional heat inputs from cooking. It is not surprising that we experience ventilation heat loss during all periods of window opening and energy requirements increase with duration of window opening.

Table 5.3: Air change rate (ACH) calculations with windows open and closed, ventilation heat loss and therefore the energy required to heat incoming air for different periods of ventilation (E_v – ventilation heat loss only).

Window position	n (ACH)	C_n (ACH)	Ventilation heat loss (W)	Energy required to heat incoming air for window opening period (kWh)				
				60 mins	40 mins	20 mins	10 mins	5 mins
Closed	2.3	2.3	293	0.293	0.193	0.097	0.050	0.023
Open	7.5	7.4	962	0.962	0.635	0.317	0.164	0.077
Increase	5.2	5.1	669	0.669	0.442	0.221	0.114	0.054

The increase in ventilation *heat loss* should also be considered alongside the *heat gain* associated with each cooking activity. The energy penalty (E_p) also takes into account heat generated by the cooking processes themselves. In this case the power outputs of the toaster and hob were 1200 W and 1700 W respectively. The power input from each cooking appliance far exceeds the measured rate of heat loss from the kitchen with the window closed at steady state of 634 W (this includes fabric and ventilation heat losses). This additional heat input from appliance use will result in a rise in kitchen temperature (under certain circumstances) and could result in overheating, necessitating the requirement for additional ventilation to maintain thermal comfort as well as good IAQ.

When we consider additional heat input from the cooking processes, we observe lower energy penalties which we attribute to lower ventilation heat losses (Table 5.4). We note that more heat can be generated from frying activities than is lost by ventilation heat loss when a window is open for a 5- or 10- minute period, so we observe an energy

surplus (shown as negative numbers in Table 5.4). In all instances of toasting we observe an energy penalty, since less energy is generated from this activity, but this is negligible for 5- and 10- minute periods of window opening. For extended periods of ventilation we lose more heat through ventilation than we gain through the cooking processes hence generate higher energy penalties. When periods of natural ventilation extend beyond 20-minutes we see larger energy penalties, even when considering the energy generated by cooking activities, due to prolonged periods of air exchange with the cooler air of the chamber.

Table 5.4: Energy Penalty (E_p) for toasting and frying per event, and for one event daily over the entire heating season (October–March).

Cooking Events	Energy penalty for window opening period (kWh)					
	60 mins	40 mins	20 mins	10 mins	5 mins	No Ventilation
One-Time (Frying)	0.531	0.303	0.082	-0.025	-0.085	-0.139
Daily over entire heating season (Toasting)	96.589	55.176	14.982	-4.507	-15.469	-25.298
One-Time (Toasting)	0.641	0.413	0.193	0.085	0.025	-0.028
Daily over entire heating season (Frying)	116.65	75.24	35.05	15.56	4.60	-5.096

To provide wider context to the findings, the increase in n of 5.2 ACH resulting from window opening was used to calculate the energy, financial, and CO_2e implications for each window opening period over an entire heating season for a similar house with gas central heating in North West England. These calculations presented in Table 5.5 and Table 5.6 are based upon the number of days in each month during the heating season (October–April) and temperature difference between the kitchen and external environment for each month assumed by SAP 2012 (Government Digital Service, 2014).

For space heating, we assumed a gas, as opposed to electric, central heating since this is the most common heating method in the UK. We assumed a heating efficiency of 82.5% based on average efficiency data provided by the Energy Savings Trust (Energy Savings Trust, 2021), which means we adjusted our energy demand accordingly. We report our results in terms of gas consumption in kWh, pounds sterling, and kilograms of CO_2 equivalents (using the BEIS methodology (UK Government, 2020) to convert gas consumption in kWh to CO_2e). Our values report the total energy penalty for one event occurring daily over the duration of the heating season. We observe modest energy penalties for both frying and toasting activities over the heating season, with slightly higher costs associated with toasting due to the larger ventilation heat losses we previously calculated. Gas consumption, financial cost and CO_2e evaluation increase with extended periods of natural ventilation (window opening). In the case of frying, owing the energy surplus we noted previously for window opening periods of 5 and 10 minutes, we see small savings. We observe more elevated financial and energy costs for enhanced periods of ventilation longer than 20 minutes.

Table 5.5: Space heating gas consumption, financial cost and CO_2e for one event per day over heating season for frying activity.

	Window opening period				
	60 mins	40 mins	20 mins	10 mins	5 mins
Gas consumption (82.5% efficient boiler)	117.077	66.880	18.159	-5.463	-18.750
Financial cost [£]	5.10	2.92	0.79	-0.24	-0.82
CO_2e (kg)	21.52	12.30	3.34	-1.00	-3.45

Table 5.6: Space heating gas consumption, financial cost and CO_2e for one event per day over heating season for toasting.

	Window opening period				
	60 mins	40 mins	20 mins	10 mins	5 mins
Gas consumption (82.5% efficient boiler)	141.40	91.20	42.48	18.86	5.57
Financial cost [£]	6.17	3.98	1.85	0.82	0.24

CO_2e (kg)	26.00	16.77	7.81	3.47	1.02

Mechanical Ventilation

In using mechanical extract ventilation, we need to consider both the energy associated with additional space energy consumption (in response to air exchange and ventilation heat loss) (Table 5.7) and the energy that goes into the extract operation. In this instance the 22.6 W power rating of the extract fan makes a small contribution to the energy penalties. The calculations in Table 5.8 include energy lost through additional space heating, energy lost through fan operation and energy gained through frying activities. Taking all these factors into account we still see minimal energy penalties which are much lower than those incurred by natural ventilation for similar ventilation periods in all events.

Table 5.7: Air change rates with extract ventilation on and off, ventilation heat loss quantified, and energy required to heat incoming air for operation period (E_v).

	n (ACH)	Ventilation heat loss (W)	Energy required to heat incoming air for extract operational period (kWh)					
			60 mins	40 mins	30 mins	20 mins	10 mins	8 mins
Extract off	2.3	293	0.293	0.193	0.146	0.097	0.050	0.038
Extract on	7.0	888	0.888	0.586	0.444	0.293	0.151	0.115
Increase	4.7	596	0.596	0.393	0.298	0.197	0.101	0.077

Table 5.8: Energy penalty (E_p) for frying per event and for one event daily over heating season.

Cooking events	Energy penalty for extract operation period (kWh)						
	60 mins	40 mins	30 mins	20 mins	10 mins	8 mins	No extract
One-time	0.480	0.270	0.171	0.063	-0.036	-0.061	-0.139
Daily over entire heating season	87.308	49.051	31.048	11.457	-6.631	-11.100	-25.298

Similar to natural ventilation, when we consider shorter (8- and 10-minute) periods of mechanical ventilation we observe an energy surplus due to additional heat generated by frying activities. When we operate the extract ventilation for periods of 20 minutes or more these costs increase, but still do not exceed those attributed to natural ventilation (window opening). We therefore see mechanical extract ventilation as the most appropriate strategy for meeting the dual objectives of good IAQ and energy efficiency.

In addition to our observation of modest energy requirements for ventilation heat loss and extract operation associated with the mechanical ventilation (Table 5.8), particularly over shorter "intermittent" periods of operation, we also incur lower space heating gas consumption requirements for the same equivalent periods of operation as window opening (natural ventilation) in all instances, again with energy surpluses when operating extract ventilation for only 8- or 10-minute periods (Table 5.9). We also observe lower or similar financial costs for an equivalent period of ventilation. This strengthens our previous assertion and leads us to re-iterate that this form of ventilation may help achieve the dual objectives of improved IAQ and energy efficiency.

We know from our PNC profiles that mechanical extract ventilation is more than capable of providing significant IAQ benefits. In fact, in the instance of frying we saw mechanical ventilation as most effective at mitigating exposure to particle number concentrations. This is despite mechanical ventilation having a slightly lower ACH than natural ventilation. However, we know the rate of ventilation is much more consistent when mechanically driven. The first 20 minutes appear to be the most significant for reducing particle concentrations. We only observe negligible energy penalties during this period. This leads us to conclude that this initial 20-minute period of extract

ventilation after cooking represents the ideal time to balance the dual objectives of good IAQ and energy efficiency.

Table 5.9: Space heating gas consumption, financial cost and CO_2e for one event per day over heating season for frying for various periods of mechanical extract ventilation.

	Extract operation period					
	60 mins	40 mins	30 mins	20 mins	10 mins	8 mins
Gas consumption (82.5% efficient boiler) [kWh]	100.842	56.165	35.140	12.802	-8.223	-13.479
Financial cost (inc. fan) [£]	5.07	2.89	1.87	0.70	-0.33	-0.58
CO_2e (inc fan) [kg]	19.6	11.0	7.0	2.6	-1.5	-2.5

It is important to consider (as we later discuss) that the energy penalties from natural ventilation may be underestimated compared to real-world situations where we would experience greater pressure and temperature differentials, whereas mechanical ventilation estimates are likely to be robust and better reflect real-world situations. Mechanical ventilation will also provide a more constant rate of air exchange than window opening, which will be more greatly influenced by external forces. We therefore assert that mechanical ventilation can be used to provide a cost-effective way to improve IAQ (and negate any negative consequences of cooking-generated pollutants) without adversely impacting upon energy efficiency.

5.4. Discussion

In the present study we observed that residential cooking, which is conducted daily in most homes, generates significant particle number concentrations (PNC). Therefore, we can see how it is regarded as a major pollutant source in residential microenvironments. Since source removal is not possible, effective removal of air contaminants generated inside residences by cooking is needed to provide good IAQ and protect the health and

safety of occupants (Singer et al., 2011). In any house with its purpose-provided openings closed and mechanical systems switched off, the total airflow rate is equal to the infiltration rate (O'Leary et al., 2019). Pollutants can be diluted by infiltration of outside air which is desirable to save energy. Whilst infiltration alone is typically not sufficient to dilute particles (O'Leary et al., 2018), the greater ACH of the "leaky" Energy House can reduce PNC to some degree, albeit not as effectively as purpose-provided ventilation, in either natural or mechanical form. When windows are opened, the air exchange rate increases from 2.3 to 7.4 ACH and we observe significant dispersion of cooking-generated particles through indoor-to-outdoor transport. Window opening has a more significant effect of reducing particle exposures for the first 20 minutes which we observe through higher initial decay rates (Appendix B).

No energy penalties are associated with non-ventilated scenarios, however, the IAQ penalty may be considerable as we have seen PNC up to 100 times greater than background concentrations during our episodic cooking events with more prolonged persistence. Some ventilation is clearly better than no ventilation from an IAQ perspective. If we exclude the energy inputs associated with cooking events, we see modest energy penalties which increase over time with extended periods of natural ventilation. If we include the heat generated through cooking activities in our calculations, then we lower the energy penalties associated with natural ventilation. There is a negligible effect on ventilation heat loss when windows are opened for short periods of time (Figure 5.7). This is particularly evident for frying activities, as a consequence of the greater heat generated during this activity. However, whilst our energy penalties are low on the order of a single experiment, they increase when we scale up to realistic values over a heating season. It is also important to note that our scaling is based on a single, short-lived cooking event per day. When cooking for longer

durations or more complex meals, we would expect different pollutant and particulate responses, and also a different corresponding energy response or implication, how negative or positive this may be, has not been quantified (Farmer et al., 2019).

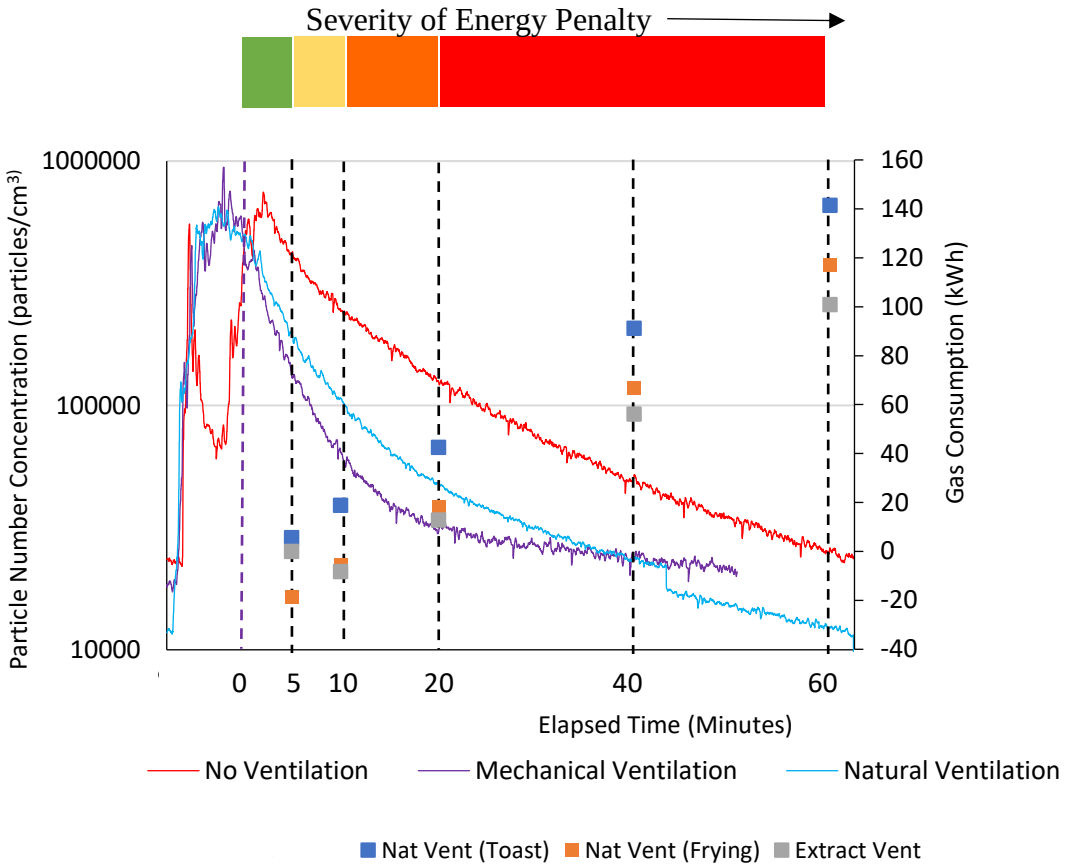


Figure 5.7: Particle number concentration averaged for all natural, mechanical and no ventilation scenarios for frying to illustrate the correlation between energy cost and implication and IAQ benefit. Gas consumption (considering space heating energy consumption and for mechanical extract ventilation, extract operation) illustrated relative to particle concentrations in squares. Severity of energy penalty is colour coded.

The amount of heat generated by cooking activities can exceed the amount of heat lost through ventilation heat loss during periods of window opening, giving an energy surplus (assuming energy is lost to the air). The energy required to fry an egg was greater than the additional energy required to heat the air exchanged with the chamber during both a 5- and 10-minute period of natural ventilation. Whilst there are no energy penalties associated with opening a window for a 5 or 10-minute period, there are

significant IAQ benefits (Figure 5.7). In terms of PNC, after peak concentrations have been reached, ventilation is most important in reducing them to lower levels through the process of enhanced air exchange. Therefore, what we have shown here is that even a ventilation period of 5 minutes can yield significant benefits in terms of IAQ improvements with no detriment to energy efficiency. This echoes a key recommendation from a study by Guyot et al. (2018) that ventilation needs be smarter, ventilating less when it does not provide an IAQ advantage. We could potentially stop ventilation after a 5- or 10- minute period having improved IAQ, without any energy penalties.

Again, for toasting activities we observe no energy penalty for no ventilation. However, the IAQ penalty would be considerable. The energy required for toasting was less than the additional heat loss even over a minimal (5-minute) ventilation period. There was no energy surplus associated with toasting activities due to the lower amount of energy generated by this activity. We observe modest energy penalties for periods of window opening, however these are minimal for 5- and 10-minute periods, suggesting natural ventilation can also be used over such timescales to improve IAQ at no real detriment to energy efficiency.

It was important to estimate an initial decay rate since this is when pollutant concentrations are highest and when a user may benefit most from using ventilation (Dobbin et al., 2018) (Appendix B). Opening a window for more than 20 minutes appears to offer fewer additional benefits in terms of improved IAQ but does result in a higher energy penalty. In our experiments we observe the same time to background when we open the window for 20 and 40 minutes and similar decay rates (particular for frying) (Appendix B). We might be observing the same overall time to background due

to initial decay rates being significantly higher for the first 20 minutes, driven initially by dispersion, then more slowly thereafter as a result of deposition and other removal processes. With regards to energy savings, we can see that opening a window for 40 minutes requires much more energy to maintain room temperature than opening a window for 20 minutes. Therefore, we conclude that there are financial and environmental benefits to closing windows 20 minutes after a cooking activity. Providing ventilation via window opening for 20 minutes after cooking can provide significant IAQ improvements without being at a significant cost to energy and we observe this delicate balance and the merging of these parameters in Figure 5.7 and see how a 20-minute period of ventilation can be complementary to both objectives. This finding is consistent with other papers that suggest between 10–15 minutes of additional ventilation following cooking practices to increase decay rates without leading to any problems associated with long-term ventilation including energy use and uncomfortable noise levels (Dobbin et al., 2018; O'Leary et al., 2019a).

Emitted pollutants can be extracted at their source using a cooker hood which reduces exposure risks by capturing emitted pollutants (O'Leary et al., 2019a) and by increasing overall air exchange in the home to remove pollutants from the indoor environment (Dobbin et al., 2018). Previous studies have shown that exhaust devices that do not cover the in-use burners suffer a large penalty in capture efficiency (O'Leary et al., 2019a). In this study, we use intermittent extract ventilation away from the stove, but still observe significant increases in air exchange (to 7.0 ACH) which promote rapid decay of PNC. This could highlight fan flow rate is more important than fan position, but we do not have comparative data to support this assertion. Regular and appropriate intermittent usage of a kitchen exhaust fan during cooking can reduce exposure to particles as we have observed in this study, but decisions about the design and use of

such fans needs to take IAQ improvements and energy costs into consideration (Rim et al., 2012).

Mechanical ventilation incurs low energy penalties. These are lower than those incurred by natural ventilation, even when the combined energy costs of extract operation and additional space heating consumption are considered. We even see an energy surplus when operated for 10 minutes or less. Energy penalties increase for periods of operation longer than 10 minutes but do not exceed those attributed to window opening in any event. We therefore suggest the use of mechanical extract ventilation over natural ventilation to better achieve the dual objectives of good IAQ and energy efficiency.

When considering energy consequences in isolation we find it would be best to operate extract ventilation for relatively short periods of time (8- or 10-minute periods), however, when we give consideration to IAQ issues we acknowledge it would be better to operate it for longer time periods to increase air changes and reduce potential exposures. Whilst we operated mechanical extract ventilation for a 30-minute period and found this to be an appropriate length of time to yield considerable air quality improvements, we know that the first 20-minutes are the most important for reducing particle concentrations. Whilst we incur only modest energy penalties for 30-minutes of extract operation, we suggest that an optimum period of 20-minutes could better balance the complimentary goals of improved IAQ and increased energy efficiency. This will yield significant improvements in IAQ with no significant or detrimental energy penalties. Mechanical ventilation also has a significant role to play in maintaining thermal comfort. We must consider its operation, particularly in colder climates, where we would experience and incur greater energy penalties and problems of thermal comfort if we were to rely solely on natural ventilation.

These findings and conclusions are similar to other papers that address IAQ concerns from cooking. Dobbin et al. (2018) and O'Leary et al. (2019a) have previously reported that exposure to UFP and $PM_{2.5}$ can be reduced by continuing to ventilate for 10–15 minutes after cooking. These papers acknowledge the trade-offs in long-term exhaust fan use, including the noise it generates and energy it uses, and acknowledge that it may be impractical to suggest residents use their exhaust fan for long periods after cooking (O'Leary et al., 2019a). Therefore, these represent optimum timings where the kitchen fan incurs only a small cost and the benefits of ventilation are considerable and the costs of ventilation (and associated heat loss) are negligible (Dobbin et al., 2018). O'Leary et al. (2019a) similarly showed daily average particle concentrations reduce by 58% for 10 minutes of extra ventilation after cooking ends when compared to ventilation during cooking alone but increasing the additional ventilation period of 15 minutes only reduces concentrations by a further 8% giving a total reduction of 66%. This is similar to our previous finding about ventilating for over 20 minutes with window opening.

It is important to consider that energy penalties of mechanical extract ventilation will increase during long-term use, though we do not yet understand what this might look like, and when scaled up to represent real-world use as we experience. However, the energy penalties of mechanical ventilation are robust and likely to reflect real-world situations, whereas those for natural ventilation may be an underestimated due to the characteristics of the chamber (and under-estimations of pressure differentials) in which the Energy House is located.

This pilot study makes key associations between IAQ and energy efficiency, but there are limitations. It is important to note that the assumptions and calculations made in this study are only applicable to houses similar to the test house under certain environmental

conditions. We cannot make generalisations for other types of housing, with regards to ventilation heat loss calculations. In the real world, we would expect that wind pressures would result in higher ventilation heat loss than we saw in our chamber experiments, particularly in winter months and under conditions of natural ventilation. We might therefore observe greater energy penalties. However, it might also be the case that the area by which a window is opened by an occupant may be dictated by external conditions (e.g., only by a small amount on a windy day).

Issues of experimental design, data collection error and fieldwork practicalities are not uncommon to studies of this type. Large scale in-situ monitoring of this type is invasive, cost and time prohibitive. One main limitation of this pilot study is the small number of cooking tests performed that limited the ability to find statistically significant differences between test conditions. The range of cooking activities is also limited. In the experimental design it was decided that mechanical ventilation would be controlled through an extractor fan mounted to the exterior kitchen wall. In reality a range hood would operate above the cooker, removing particles before they disperse with the kitchen air, so we would expect lower PNC.

The insights provided in this study are restricted to wintertime in the UK and single-order experiments. Whilst we did not consider seasonal variations, we can make inferences based on temperature changes. We would expect increased air exchanges in the summer due to frequent window opening, which would drive faster decay rates. Higher outside temperatures would also result in lower energy penalties with less need for any temperature recovery. Further work could quantitatively examine the effect of seasonality on energy penalties to exposure mitigation. We could also think of taking a similar approach to cooking as HOMEChem and examine the energy consequences of

layered or more complex cooking events (Farmer et al., 2019). We have introduced a method that could be adapted for more complex cooking events and the energy consequences we observe could be very different as a result. For more prolonged cooking events or for a mixture of food types, we might expect prolonged pollutant profiles, and enhanced pollution, and therefore might need a more prolonged ventilation strategy. This may be counteracted in part by enhanced heat generated by the cooking processes.

5.5. Conclusions

This pilot study brings new data on dealing with the -sometimes- conflicting objectives of good IAQ and energy efficiency in existing dwellings. Through examining temporal indoor pollutant concentrations from discontinuous cooking activities, we examined the energy penalties of ventilation, that is the additional energy consumption resulting from actions undertaken to reduce the concentration of cooking generated pollutants. We highlight how the apparently conflicting objectives of good indoor air quality and energy efficiency can be achieved through, most importantly, intermittent mechanical ventilation in this type of house. We highlight how natural ventilation can also be important, particularly for the first 20 minutes after cooking in this study, despite incurring slightly higher energy penalties.

We do not want gains in IAQ to be made at the expense of energy efficiency and our study has generally shown that for short periods of time (up to 20 minutes) this is generally not the case. Considering both IAQ and energy efficiency, we conclude that ventilating for 20 minutes after a cooking event can yield significant IAQ benefits with negligible energy penalties. Ventilation for an additional 20 minutes appears appropriate and may be memorable as it is simple, which could make it a potentially

suitable guideline. With these conclusions in mind, it is important to consider that the choice of ventilation and its use over time depends on the behaviour of the occupant, and modification of occupant behaviour will be important moving forward. The implications of this study are significant in the context of residential energy use and plans to meet UK energy reduction targets by 2050 (Eyre and Baruah., 2015).

6.0. Does BREEAM Accreditation Reflect Good Indoor Air Quality?

An Exploratory Study Quantifying VOCs within Sustainable Buildings

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Abstract

BREEAM is the world's leading sustainability assessment method for buildings. Indoor air quality (IAQ) is one component evaluated in BREEAM assessment, however only a few credits relate to IAQ, and emission levels of volatile organic compounds (VOCs). This study investigated the prevalence and concentration of volatile organic compounds at a medium sized British university within BREEAM certified buildings using a sieve mapping approach that involved sampling over occupied (activity-related) and non-occupied (building-related) time periods. The aim was to evaluate sources of VOCs and the relevance of IAQ credits in BREEAM accreditation in order to determine whether the current accreditation approach is fit-for-purpose and a reflection of IAQ.

Low VOC concentrations were observed, below stated guideline values and BREEAM limits. Buildings housing laboratories generally experienced higher volatile organic compound concentrations, particularly the Chemistry building where synthetic chemicals and solvents were being used. Relatively higher benzene, toluene, ethylbenzene, xylene, and monoterpene concentrations in locations adjacent to

laboratories in the Chemistry building suggests enhanced dilution of building and cleaning-related contaminants by local laboratory exhaust systems rather than passive transfer from the laboratory as we would have observed other quantified volatile compounds at higher concentrations. The most prevalent carbonyl compounds appeared to be weakly correlated to recent refurbishments within the Physics building. This study provided insights into the prevalence and concentration of volatile organic compounds in a multi-functional building environment and a solid foundation for further work quantifying VOCs within indoor environments.

The lack of association between a building's chemical footprint (VOC concentrations) and BREEAM certification leads us to question the value of IAQ credits in BREEAM standards. The novelty of this research lies in the suggestions made to develop the IAQ credits to provide a more relevant assessment of IAQ within buildings. This includes the recommendation for a new approach to quantifying VOCs in sustainable building assessments that involves simultaneous temporal measurements with sensitive TVOC detectors alongside detailed speciation with reference instrumentation over longer time scales and occupied periods.

Keywords: indoor environments, sustainable buildings, BREEAM, indoor air quality, volatile organic compounds, BTEX

6.1. Introduction

The UK population spends on average 80–90% of their time inside buildings (Dimitroulopoulou et al., 2017), where they are exposed to many pollutants. Whilst indoor air quality (IAQ) is less well characterised and understood than ambient air quality, it has become an emerging focus of research since the built environment is an important determinant of population health. It is well understood that indoor pollutants

act as respiratory irritants, toxicants, adjuvants or carriers of allergen and can lead to lung cancer, chronic obstructive pulmonary and cardiovascular disease (Spinazze et al., 2019). The effects of poor IAQ on performance, productivity, and well-being of occupants have also been studied (Wargocki and Wyon, 2017; Eitland et al., 2018; Gupta et al., 2018). The existence of a wide range of indoor micro-environments, each impacted by one or more pollutant source, complicates exposure studies (Kim et al., 2001). Whilst there is a plethora of indoor pollutants; the current work focusses on volatile organic compounds (VOCs thereafter).

VOCs are a main category of indoor air pollutant (Goodman et al., 2017). Documented sources include consumer products (air fresheners, cleaning supplies and personal care products are associated with emissions of limonene, α-pinene, acetone, acetaldehyde and ethanol) and building materials; wood (MDFs and particle boards), thermal and acoustic insulations, carpets, paints, coatings, industrial solvents, adhesives, fireproof materials, PVC, flooring and furnishings (Shaw et al., 2005; Cacho et al., 2013; Yurdakul et al., 2017; Goodman et al., 2018). Even environmentally friendly materials (also known as “green” building materials) can emit potentially hazardous VOCs. Formaldehyde, (a known human carcinogen) whose indoor concentrations typically exceeds outdoor concentrations, is often treated separately as it is not detected by the gas chromatographic methods typically used to quantify VOCs (Shaw et al., 2005). Sources include the degradation of resins and additives used in wood-based building materials, furniture, sealants and combustion and chemical reactions (Destaillats et al., 2006; Singer et al., 2006; Kruza and Carslaw, 2019). Many of these sources are pervasive in new buildings (Verriele et al., 2016). The concentration of individual VOCs in the indoor environment is dependent on their emission rates from various indoor sources, the rate at which they are transported from outdoors to indoors and the

rate at which they are scavenged by indoor surfaces, subjected to chemical and photodegradation and removed by ventilation or filtration (Weschler, 2009). Given the plentiful sources, VOC levels are usually 2–5 times higher in indoor air than in ambient air, but during certain activities can even be 1000 times higher (EPA, 2017a). If there were no indoor sources of VOCs their concentration would be lower than outdoor concentrations as VOCs are expected to adsorb on surfaces, and some are chemically degraded (Yurdakul et al., 2017).

Many VOCs frequently detected in indoor environments are associated with acute and chronic health effects such as sensory and skin irritation, headaches, breathing difficulties, asthma and cancer (Goodman et al., 2017). In the United Kingdom (UK), there are currently no enforced IAQ limits for individual VOCs. A statement issued by Public Health England in 2019 presents IAQ guidelines derived from scientific literature for selected VOCs to control their levels in the indoor environment through informing discussions on source control and raising awareness (Public Health England, 2019). Since many people are exposed to various substances at work, some of which are potentially harmful, indicative occupational health exposure limit values (IOELVs) have been introduced under the Chemical Agents Directive (98/24/EC) through Workplace Exposure Limits (WELs) (Health and Safety Executive, 2018). Exposure concentrations should be placed in context with these guidelines.

There has been a significant shift towards energy efficient construction and buildings in recent years which has led to building codes that target energy conservation and resulted in the tightening of building envelopes to reduce air infiltration and leakage (Mudarri, 2010). Recent decades have seen the introduction and growth in use of green building certification schemes which aim to promote the development of healthy,

energy saving and environmentally friendly sustainable buildings (Wei et al., 2015). The earliest certification scheme, BREEAM (Building Research Establishment Environmental Assessment Methodology), was established in the UK in 1990 and is the world's leading sustainability assessment method for assessing, rating and certifying the sustainability of buildings. Rating systems are universally used in sustainable certification schemes; whereby the requirements are divided into several categories and a scoring system is used to evaluate to what extent individual attributes within each category are achieved (Wei et al., 2015). Categories evaluated in the BREEAM assessment include energy and water use, health and wellbeing, pollution, transport, materials, waste, ecology, and management processes.

Since the introduction of these benchmarking schemes, IAQ has been included as a default element, though, on average contributes only 7.5% to the credit total (Steinemann et al., 2017; Zhong et al., 2017). IAQ is included in the health and wellbeing category in BREEAM. To gain any of the IAQ points in a BREEAM assessment, certain testing and performance requirements must be complied with (e.g., EU Directive 2004/42/CE ("Paints Directive")), TVOC and formaldehyde levels must meet certain requirements post construction (pre-occupancy) and manufacturers must confirm certain requirements such as the absence of prohibited wood preservatives/biocides (BREEAM, 2016). Points are also awarded for an IAQ plan and design and implementation of ventilation. In general, occupants of green buildings rate IAQ higher than occupants in conventional buildings (Steinemann et al., 2017). Some studies, however, highlight no observed effect of certification on the rating of IAQ.

In parallel with green building rating systems, newer designs, construction practices and building materials and the use of environmentally friendly products offer the potential

of lowering chemical exposure (Zhong et al., 2017). Supported research shows low VOC emitting building materials and consumer products can reduce VOC concentrations, and certification schemes provide credits for the use of low-emitting materials (Zhong et al., 2017). However, even though some products are marketed as green, this does not guarantee healthier products or better IAQ. Recent studies have found that products with green credentials can emit hazardous compounds, sometimes comparable to their conventional counterparts (Steinemnan et al., 2017). For example, green cleaning products often contain fragrance chemicals e.g., terpenes, that are primary and secondary pollutants (Steinemann et al., 2017). Even zero or low-VOC paints can still emit VOCs similar to regular paints since the paint tinting process can sometimes add some VOCs, as well as other problematic chemicals such as semi-volatile organic compounds (SVOCs; such as perfluorinated alkyl substances (PFASs) and siloxanes) (Steinemann et al., 2017).

Exposure to VOCs has been an important research issue because of the prevalence of these compounds in various micro-environments and their associated adverse health effects (Jo et al., 2010). Educational institutions are one of the most studied indoor environments with a focus on primary and high schools as they house high density populations and the young are considered particularly vulnerable in terms of exposure to pollutants (Akal et al., 2015; Goodman et al., 2018; Godwin and Batterman, 2007; Kolarik et al., 2015; Yurdakul et al., 2017). Zhong et al. (2017) examined VOCs in 144 classrooms in 37 conventional and sustainable US elementary schools. Most VOCs had a mean concentration below 5 $\mu g/m^3$ and the most prevalent VOCs were aromatic compounds including toluene, benzene, m/p-xylene and 1,2,4-trimethylbenzene (Zhong et al., 2017). Overall, no major differences in VOC concentrations were found between conventional buildings and buildings with sustainability credentials (Zhong et al., 2017).

Good IAQ across universities and university campuses is important due to the long time spent by staff and students in these buildings. Campus sustainability has recently become an important issue as many universities foster the development "green-campuses" and sustainable concepts (Park et al., 2014). Several universities have begun to construct new buildings and renovate old buildings to attain sustainability certification to reduce energy consumption and improve the environment and health and well-being of employees and students (Zhong et al., 2017), but there are questions around the impact of sustainability certification on IAQ. Several international studies have examined indoor air pollution in university buildings (Yurdakul et al., 2017) consistently finding indoor environments in university settings may be important sources of pollutants (Goodman et al., 2018).

Chan et al. (2007) quantified VOCs within classrooms, offices, canteens, workshops, laboratories and a library on a university campus in Hong Kong. The main VOCs detected were toluene and benzene, which were attributed to ingress from outdoors. Solomon et al. (2008) measured IAQ within the Department of Physics and Electrical Engineering at the University of Bremen. Whilst tobacco smoke was seen to be a dominant factor in indoor pollution, pollutants commonly associated with cleaning products and materials exhibited higher indoor than outdoor concentrations (Solomon et al., 2008). Similarly, Goodman et al. (2018) conducted novel research into the prevalence and concentration of VOCs at an Australian University within campus services, restrooms, renovated offices, a green building, meeting areas and classrooms. Analysis of 41 VOCs across 20 locations revealed higher indoor concentrations than outdoors. The most prevalent VOCs (e.g., ethanol, d-limonene and formaldehyde) were found to have links with building materials, furnishings and fragranced consumer products such as air fresheners and cleaning supplies (Goodman et al., 2018). Tang et

al. (2016) examined the full spectrum of VOCs emitted indoors in a university classroom and found human occupants were the major contributor to the mass of indoor VOCs (by mass, 57%). Among the most abundant species detected were compounds associated with personal care products but human metabolic emissions such as isoprene, methanol, acetone and acetic acid were also prominent (Tang et al., 2016).

On-campus chemical use is a major challenge for university environment management programmes. Most science and engineering laboratory activities necessitate the daily use of various types and amounts of chemicals including acidic and basic materials (Park et al., 2014). In laboratories specific pollutant concentrations may be high depending on the nature of the experiments conducted and number of people working; according to cited literature, due to the use of volatile solvents and chemicals, higher VOC levels have been detected in most university buildings housing laboratories (Park et al., 2014). Pollution in (university) laboratories is particularly important to those who work there including technicians, specialists and teaching/research assistants and students who may be exposed to pollutants which may adversely affect health (Ugranli et al., 2015). IAQ and occupational safety guidelines may be used to evaluate IAQ within laboratories as they are considered an occupational micro-environment for those who work there and a general micro-environment for students (Urganli et al., 2015). Most chemicals are handled in fume hoods installed as a part of the local exhaust ventilation system to protect the researchers' health and prevent deterioration of IAQ however these are adequate only for odoriferous and volatile chemicals in the restricted area of their enclosure and have been seen as a potential outdoor pollution source (Park et al., 2014).

It is important to understand the spatial distribution of VOCs within any building as this is a reflection of their indoor sources (Yurdakul et al., 2017). Spatial variations have been seen to be significant. Limited investigations involve (university) laboratories and examine the effect of chemicals and associated VOCs on IAQ. Park et al. (2014) investigated IAQ in university laboratories, noting the concentrations of 11 VOCs were significantly higher within buildings housing laboratories (Mean: 185 $\mu g/m^3$) than those that did not (Mean: 12 $\mu g/m^3$) owing to frequently used laboratory chemicals. Rumchev et al. (2003) and Valavanidis and Vatista (2006) investigated IAQ in university laboratories in Western Australia and Athens respectively, showing occupants can be exposed to higher particle concentrations and TVOC levels but reported that air conditioning reduced VOC concentrations.

The lack of research discussing the distribution patterns of VOCs within sustainably accredited buildings is concerning. To remedy this and address the knowledge gap between green building practice and potential, this study aimed to quantify VOCs between and within sustainably accredited (BREEAM) buildings. A university provides a useful mix of building ages, uses and accreditations for this study. Through examining the prevalence and concentration of numerous pollutants, with a focus on BTEX, terpenes and carbonyl compounds between buildings, this work will contribute insights on university indoor environments in the UK. This study will adopt a sieve mapping approach, starting broad and refining during the course of investigation. Based on interpretation of initial results, sampling locations were refined to the building and room scale. In particular this work builds on that undertaken by Rumchev et al. (2003) which assessed IAQ in university laboratories to assess the implications of chemical use on the building. In assessing the relationship between VOC concentrations (and sources) and BREEAM accreditation this study aimed to evaluate the relevance of IAQ credits

in BREEAM accreditation and whether the current approach is fit for assessing the sustainability of buildings and reflecting good IAQ and positive health related outcomes.

The following objectives are identified for this study:

1. Determine the concentration of VOCs for selected buildings at a UK University (sustainably accredited BREEAM buildings and otherwise) to assess the association between VOCs and BREEAM standards
2. Determine concentration of VOCs and potential VOC sources (activity-related and building-related) at a building level (multiple buildings) and a room level (multiple spaces within one building)
3. Assess the relevance and validity of IAQ credits in BREEAM standards
4. Assess the implications of the results, and potential future monitoring or BREEAM modifications

6.2. Methodology

6.2.1. Study Location

This study was undertaken at Lancaster University, a medium sized public research university in the City of Lancaster, Lancashire, England. The university was established by Royal Charter in 1964 and currently has a student population of around 14,000 and a staff population of around 5,000. The Bailrigg Campus is 360-acres and is located 3 miles south of the City of Lancaster.

6.2.2. Study Design

6.2.2.1. Selection and Characterisation of University Buildings

All of the buildings investigated in this study are used for administrative, research, or teaching purposes (Appendix C). The sample of buildings chosen were taken as representative of the wider university building stock. Most of the buildings chosen have BREEAM certification however two conventional non-BREEAM certified buildings were also used in this study for comparative purposes. The building typologies differed greatly but there are similarities in characteristics (Table 6.1). The eight buildings with various BREEAM certifications were built between 1960 and 2018. Some of these buildings are refurbished (3 buildings), and some are new builds (5 buildings). These BREEAM certified buildings were constructed with sustainable designs, featuring high standards of thermal and mechanical ventilation influenced by BREEAM Excellent and Very Good principles. Many of the buildings make use of natural light and natural ventilation. The two conventionally designed buildings which did not have any BREEAM certification were built in the late 1960s. These buildings were also mechanically ventilated.

Table 6.1: Typology for each Lancaster University building that has been sampled in this study; year built, refurbishment; BREEAM status, and presence of laboratories.

N	Name	Built	Refurbished	BREEAM	Labs
1	Physics	1964	Y	Y	Y
2	Chemistry	1964	Y	Y	Y
3	Engineering	2011	N/A	Y	Y
4	LEC	2006	N/A	Y	Y
5	FST	1964	Y	N	N
6	Management	1964	Y	N	N
7	Infolab	2010	N/A	Y	N
8	Charles Carter	2011	N/A	Y	N
9	LICA	2010	N/A	Y	N
10	Faraday	1964	Y	Y	N

6.2.2.2. Sampling Periods, Sites and Sample Collection

Time-integrated samples were collected through active sampling over several non-consecutive days in 2018/2019. A sieve mapping approach to sampling was adopted, starting off with broad sampling in a range of university buildings, followed up by refined sampling in locations or hotspots of interest. University-wide VOCs sampling was initially undertaken during periods of low-occupancy, during university vacation periods to place a larger emphasis on assessing the influence of the building fabric on IAQ. Subsequent sampling was undertaken during periods of higher occupancy (term time) in order to place emphasis on capturing pollutant-generating activities as well. For VOC sampling (other than carbonyl compounds) a multi absorbent tube (Markes Tenax/Carbograph) was connected to a SKC Pocket Pump 210 (Eighty Four PA, USA) initially at a flow rate of 500 ml/min for 20 minutes (10 L) and then for subsequent sampling at a flow rate of 300 ml/min for 15 minutes (4.5 L) which is within the range recommended by HSE (Health and Safety Executive, 2016).

For carbonyl compounds, the sampling protocol was the same as for VOCs with regard to a broad approach across multiple university buildings, and then refined sampling in locations of interest. Air was sampled onto 2,4-dinitrophenylhydrazine (DNPH)-treated silica cartridges from stable derivatives in situ, connected to an SKC Pocket Pump at a flow rate of 500 ml/min for 4 hours (120L). Sampling took place over weekends during periods of low occupancy from 8am–12pm and 12pm–4pm each day. An ozone scrubber was placed in front of the cartridge to prevent ozone interference. After exposure the glass tubes were sealed. The methods used for analysing VOCs and carbonyls are consistent with international protocols (e.g., U.S. EPA Methods TO-17).

During university-wide sampling in periods of high and low occupancy, indoor samplers were set up in open access locations within the foyers of buildings (free from

significant polluting activities, and consistent across all buildings). Throughout all of the sampling, samplers were set at approximately 1 m in height and away from windows, doors and potential emission sources (where possible during refined sampling), out of direct sunlight and at least 0.5 m away from bookshelves and other potentially stagnant areas. Locations were also chosen that were secure from tampering. This protocol is the same as used by Godwin and Batterman (2007).

Following initial campus-wide sampling, higher resolution sampling was undertaken in "hotspots" of activity. Two buildings were selected for this purpose. With regards to VOC sampling, the Chemistry building was revisited three further times for sampling (Figure 6.1) during occupied periods to capture activity related influences. This building was selected for further analysis based on initial results of elevated concentrations, and because of the volatile and carcinogenic chemicals used by staff and students in the laboratories. Laboratories are also situated close to classrooms and personnel offices, and hence there is obvious interaction and possible transference. We specifically intended to investigate this. For carbonyl compounds, based on initial results, samples were taken at a higher spatial resolution within the Physics Building (Figure 6.2) during an unoccupied period to place larger emphasis on capturing building related influences.

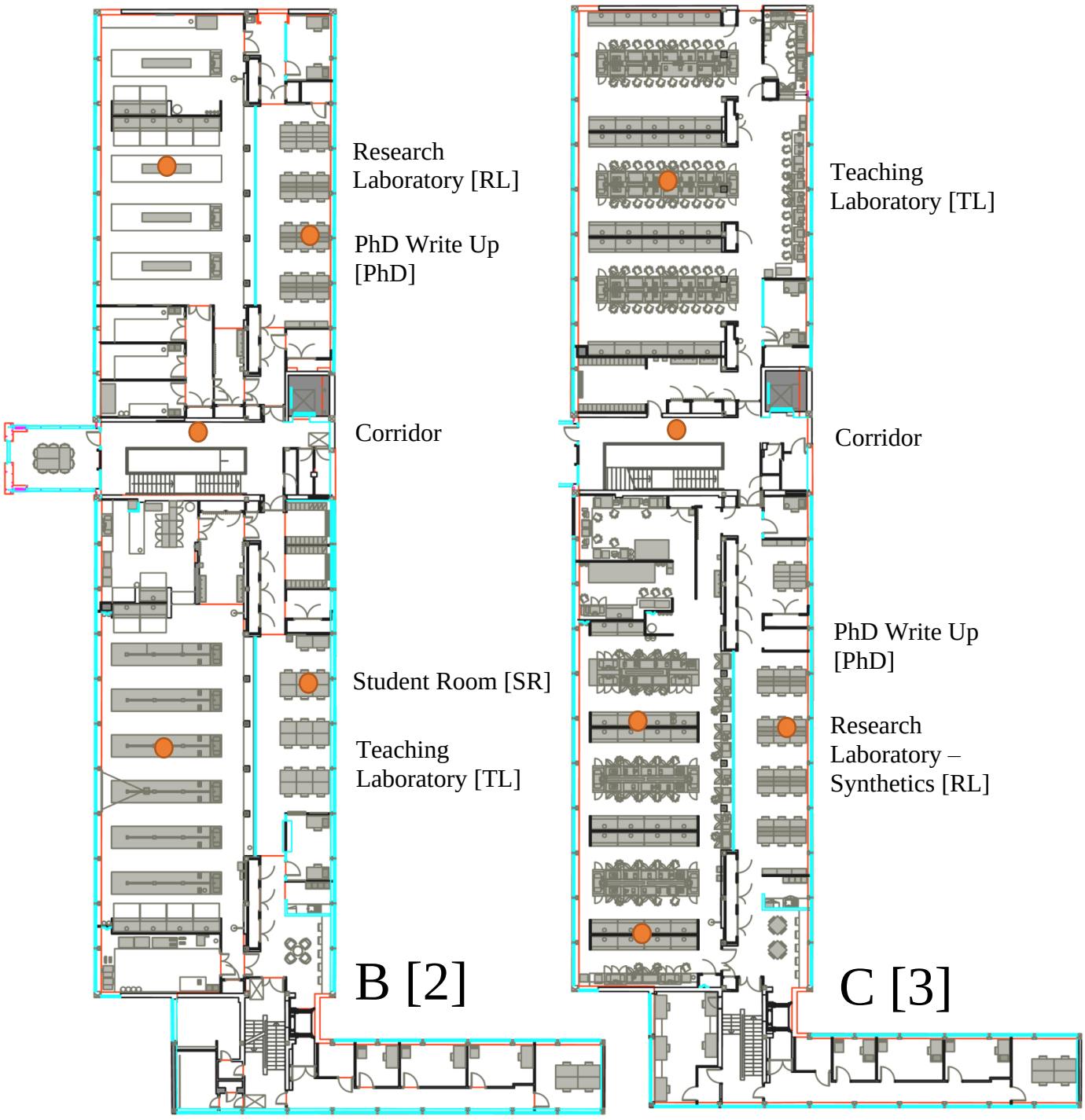


Figure 6.1: Floor Plans of the Chemistry Building [Floor B [2] & Floor C [3]]. Floor plans are scaled and edited based upon CAD drawings obtained from the University. The locations of samples from Survey 1 & 2 are illustrated with orange circles. The following acronyms are used throughout the paper: RL – Research Laboratory, TL – Teaching Laboratory, SR – Student Room, PhD – PhD Working Room and Floor Number illustrated by [2], B Floor and [3], C Floor.

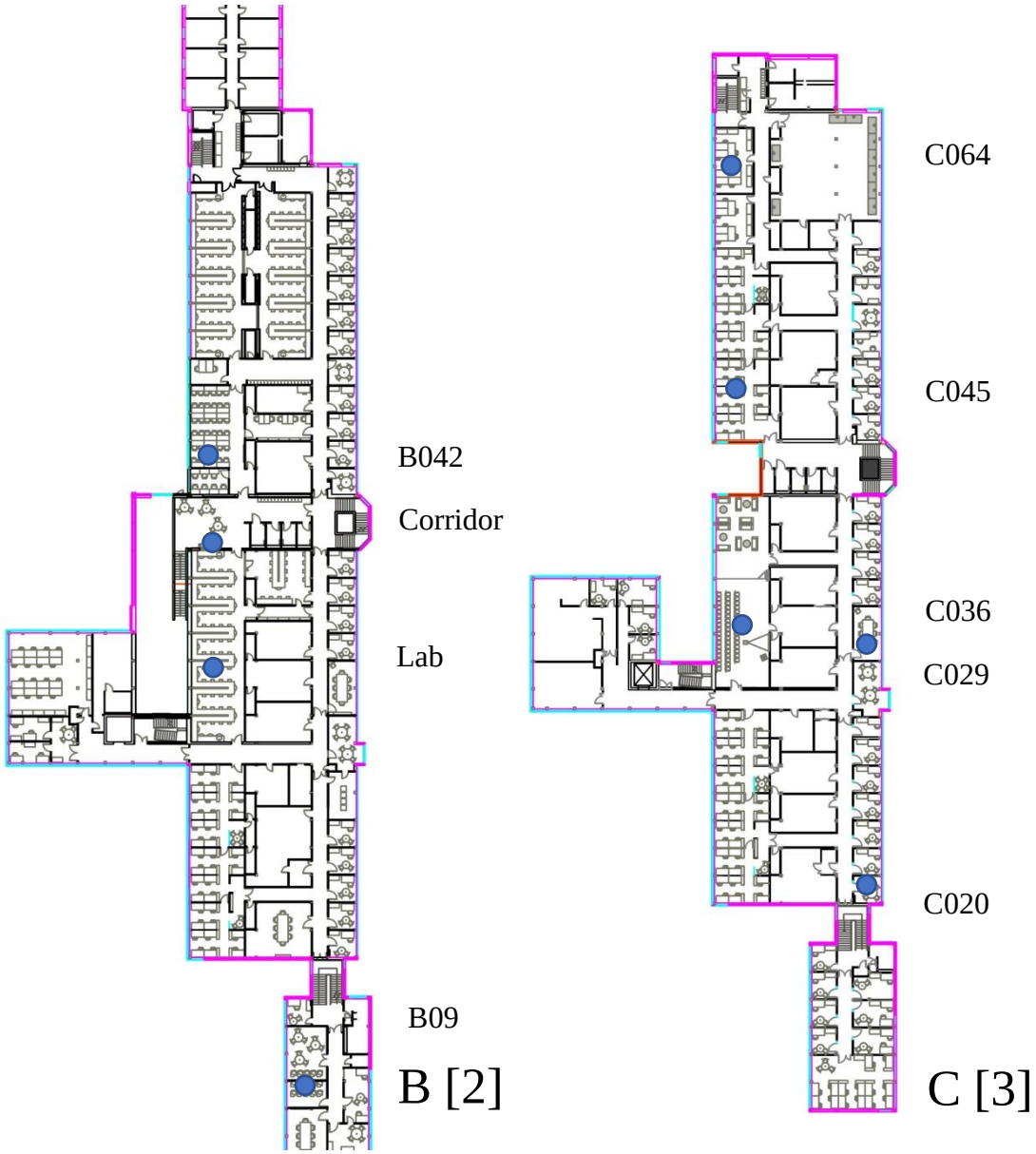


Figure 6.2: Floor Plans of the Physics Building [Floor B [2] and Floor C [3]]. Floor plans are scaled and edited based upon CAD drawings obtained from the University. The locations of samplers are shown with blue circles. Room numbers of these locations are also given.

6.2.2.3. Analytical Methods

A total of 40 VOC tubes and 20 carbonyl cartridges were analysed. For quantifying individual species of VOC, thermal desorption tubes were analysed using a Markes automated thermal desorber (ATD) TD100-xr and an Agilent 7890A gas chromatography and Agilent 5975C mass spectrometer. This thermal desorber involved

sample desorption and transport into the GC column. An Agilent DB VRX fused silica capillary column was used for compound separation. The procedure was developed in house at Marchwood Scientific.

For comparative purposes we focused on analysis of the most prevalent and most extensively studied VOCs including BTEX and monoterpenes (Verriele et al., 2016) so these compounds were fully quantified in all samples. The same compounds were quantified in a similar study undertaken at the University of Birmingham (Kim et al., 2001). Certified gas standards were used for this external calibration, including benzene, toluene, ethylbenzene, and xylene (BTEX) standards and standards for monoterpenes. For semi-quantification of the top 10 most prevalent compounds in each sample, deuterium substituted hydrocarbons were used as internal standards. The mass spectrometry response of these substances in each sample is used to quantify what is observed in the sample itself. To determine blank values, a non-exposed cartridge was analysed for each batch of samples.

For the analysis of carbonyls (which in this study included formaldehyde, acetaldehyde, propionaldehyde, crotonaldehyde, butylaldehyde, benzaldehyde, isovaleraldehyde, valeraldehyde, hexaldehyde and 2,5-dimehtylbenzaldehyde) high performance liquid chromatography with ultra-violet detection (HPLC-UV) was used. An Agilent 1100/1200 HPLC system was used in conjunction with a C-18 reverse phase column and UV/diode array detector. This method has been validated based on standard protocols.

6.3. Results and Interpretation

6.3.1. Concentration and Prevalence of Compounds between Buildings

Overall, we observe good indoor air quality (IAQ) across the range of buildings monitored, which we largely attribute to relatively high air exchange rates owing to mechanical ventilation. Full quantification of BTEX and monoterpene compounds in initial samples from across the University revealed that for many compounds concentrations were close to limit of detection (LOD) of 0.005 µg/m^3 although relatively higher individual and total VOC levels were found in buildings housing laboratories; Chemistry (7.1 µg/m^3), Physics (3.8 µg/m^3) and LEC (4.5 µg/m^3) compared to other non-laboratory buildings (Figure 6.3). Heightened VOC levels thus positively correlate with buildings where strong VOC emission sources are most likely present. This finding is consistent with the findings of Park et al. (2014) and Yurdakul et al. (2017) who found higher VOC concentrations in university buildings housing laboratories due to the use of volatile solvents and chemicals. VOC concentrations in each building housing a laboratory differed from one another because of the unique experiments conducted and materials used within each building. Since samples were taken from the foyer of each building the results suggest that volatile chemicals from laboratories influence the air in other areas of these buildings (Valavanidis and Vatistas., 2006).

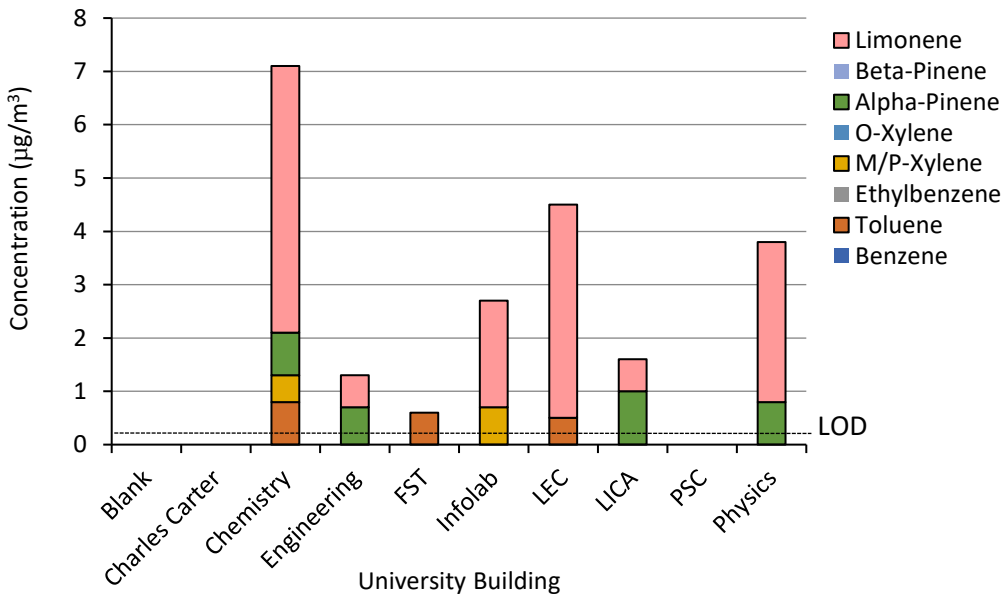


Figure 6.3: Quantification of BTEX, and mono-terpene compounds across university buildings (ordered alphabetically). *All buildings BREEAM-certified with the exception of FST (Appendix C1). **Based on 1 20-minute "grab" sample at a flow rate of 500 ml/min per building, that being a single sample or measurement taken at a specific time or over as short a period as feasible**,** taken August 2018.

The monoterpenes; limonene and α-pinene were the most prevalent compounds in the samples taken across the university campus. Wang et al. (2017) also saw higher variability in monoterpene concentrations compared to other species. The strong presence of monoterpene compounds likely reflects cleaning activities. Previous studies (Nazaroff, 2004) attribute the presence of these compounds to fragranced consumer products and cleaning products, so it not unusual to expect them to be prevalent here. Additional analysis highlighted there were other terpene compounds found at similar concentrations to limonene and pinene as well as significant levels of isopropyl alcohol (60 $\mu g/m^3$) in the sample from the Infolab which is unexplained. Toluene, a component of household products and used in renovation activities is present in several of the samples, though at low concentrations which is not unexpected. A higher level of toluene in science buildings or buildings housing laboratories is not uncommon as it used as a solvent. Since the samples are taken in the entrance foyer, they are unlikely to have been influenced by smoking due to the smoke-free workplaces and public spaces around the university. Toluene and monoterpenes have similarly been the identified as the most prevalent VOCs in numerous studies (Godwin and Batterman, 2007).

Relative to other buildings, higher levels of most individual VOC species were observed in the Chemistry building. Again, this is not unexpected since the laboratory environments in the Chemistry building are likely to provide strong sources of VOC emissions. Activities here play a crucial role in VOC emissions especially the storage, use and application of chemicals and solvents. The presence of these compounds could

highlight the influence of laboratory environments on the IAQ of the building as a whole since the sample was taken from the foyer, but there may also be a VOC influence from the building fabric. Whilst this sample is considered representative of the building as a whole and we can see that it likely reflects activities within the building, it does not reflect potential spatial variability within the building. As relative VOC concentrations were higher in Chemistry than other buildings, this prompted further detailed investigation (section 6.3.2).

Common aldehydes were quantified in similar locations to traditional VOCs (Figure 6.4). All aldehyde concentrations were well below health and occupational exposure limits. Although there are no direct occupational health exposure limits based on the duration of sampling (4 hours), much lower formaldehyde and acetaldehyde levels were observed than prescribed by WELS long-term exposure limits (8 hours).

The most prevalent aldehyde was formaldehyde. The prevalence of carbonyl compounds in general and formaldehyde in particular is confirmed in the literature by Geiss et al. (2011) who measured high concentrations for these species in public buildings and schools (Verriele et al., 2016). Formaldehyde is ubiquitous and can be emitted from a range of building, furnishing and decoration materials (Singer et al., 2006; Kruza and Carslaw, 2018) and has been identified in numerous indoor environments. Indoor formaldehyde levels have been found to be 2–3 times higher in newly built schools than existing buildings (Verriele et al., 2016). This statement holds partially true in this study. Newer or newly refurbished buildings (Physics (42 $\mu g/m^3$), LICA (13 $\mu g/m^3$), Charles Carter (14 $\mu g/m^3$) and LEC (13 $\mu g/m^3$)) have elevated formaldehyde concentrations. Newer furniture and construction materials are postulated to be responsible for elevated concentrations. Recent refurbishment (2016) of the

Physics building prior to the sampling period (2018/2019) is likely to have resulted in elevated concentrations and a maximum formaldehyde concentration of 42 µg/m^3.

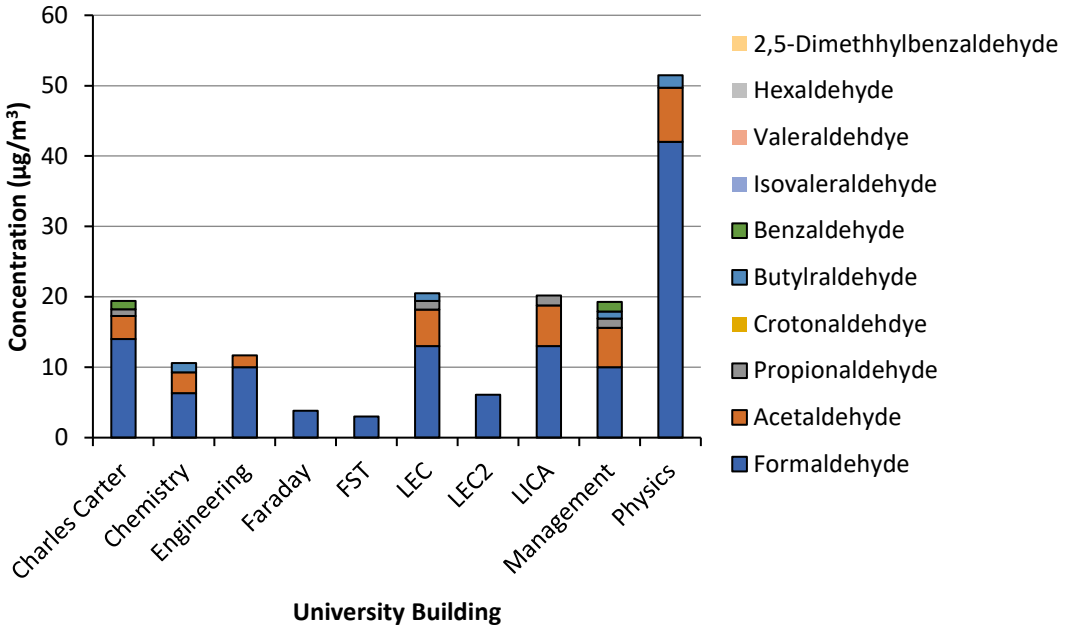


Figure 6.4: Concentrations of aldehyde compounds from samples taken within buildings on Lancaster University campus. *All buildings BREEAM-certified with the exception of FST and Management. **Data based on 1 4-hour "grab" sample at 500 ml/min per building, and measurements taken February 2019.

Higher levels of aldehyde compounds (including commonly identified formaldehyde and acetaldehyde) have been related to pressed wood products, urea-formaldehyde foam insulation, particle-board furniture and interior finishing materials and surface coverings of walls, floors and ceilings, and other textiles and adhesives (Derbez et al., 2014). Although the actual sources of aldehyde compounds cannot be conclusively identified due to the vast array of potential sources, most buildings have plenty of materials, plastic and foam furnishing which can slowly emit these chemicals overtime. When averaging concentrations, we observe greater aldehyde concentrations in sustainably accredited BREEAM certified buildings than non-BREEAM certified buildings. This could be attributed to the greater number of green certified buildings monitored. However, we see that even in green buildings, where construction and

finishing materials are selected based on performance and environmental parameters, the highest formaldehyde and acetaldehyde concentrations have been found to be associated with compressed wood products, wood finishing materials, adhesives and occupant density (Goodman et al., 2017).

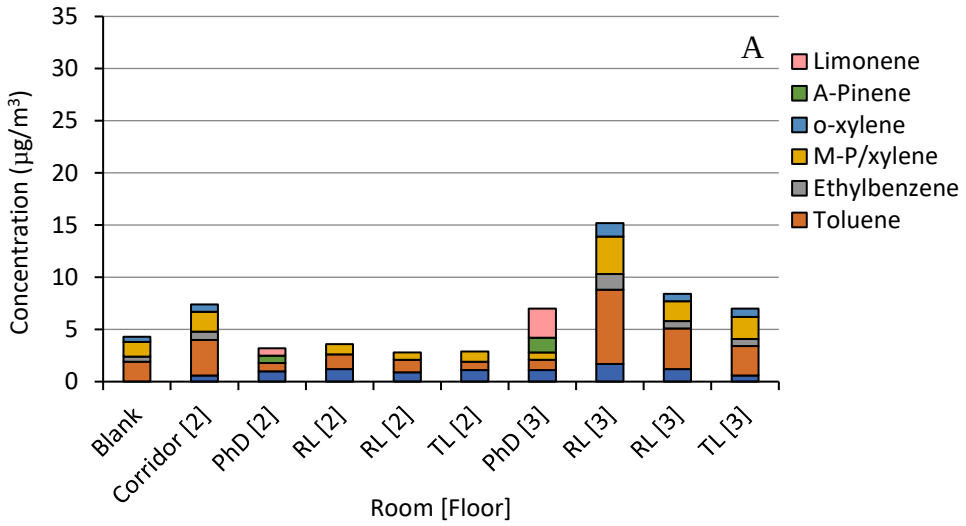
The results from this study are consistent with the findings in previous studies (Kaden et al., 2010). In a prior study at an Australian University formaldehyde had geometric mean concentrations in classrooms (16.9 μg/m^3), renovated offices (14.2 μg/m^3), and a green building (13.6 μg/m^3) with a range in other locations from 4.5–7.2 μg/m^3 (Goodman et al., 2018). These levels are within the magnitude of those observed in the present study across all building types. At a new university campus located in China, the average formaldehyde concentration in teaching buildings was 46.0 μg/m^3 (Kang et al., 2017). The average formaldehyde concentration for similar spaces in this study was 17.8 μg/m^3. Higher levels in the Chinese study when compared to our own may be partly attributed to the more recent construction of the university. Other studies have shown higher VOC levels typically occur post-renovation or in the first few years post-construction.

6.3.2. Concentration and Prevalence of Compounds within Buildings

6.3.2.1. Chemistry Building

Results highlight comparatively higher concentrations of BTEX and monoterpene compounds outside of laboratory environments in some locations (RL [2] vs PhD [2]) but not in others (PhD [3] vs RL [3]) in the first survey (Figure 6.5a). Results in the following surveys were similar though concentrations of BTEX and monoterpene compounds were generally higher, despite a lower sampling period and sampling volume, and spatial patterns clearly emerged, with significantly higher concentrations

outside of the laboratory (PhD [2] vs RL [2], SR [2] vs TL [2] and PhD [3] vs RL [3]) (Figure 6.5b). In general, these concentrations are higher than observed previously within this study with the exception of limonene which generally exhibits lower concentrations suggesting the presence of strong emission sources or activities. Higher BTEX and monoterpene concentrations outside of laboratory environments in the Chemistry building suggest some passive transfer of chemicals from the laboratories into nearby and adjoining environments. Whilst these chemicals are also associated with building materials and cleaning products which are detected at relatively low levels throughout the rest of the building, higher concentrations here suggest they originated in the laboratory where there are numerous additional sources related to ongoing activities.



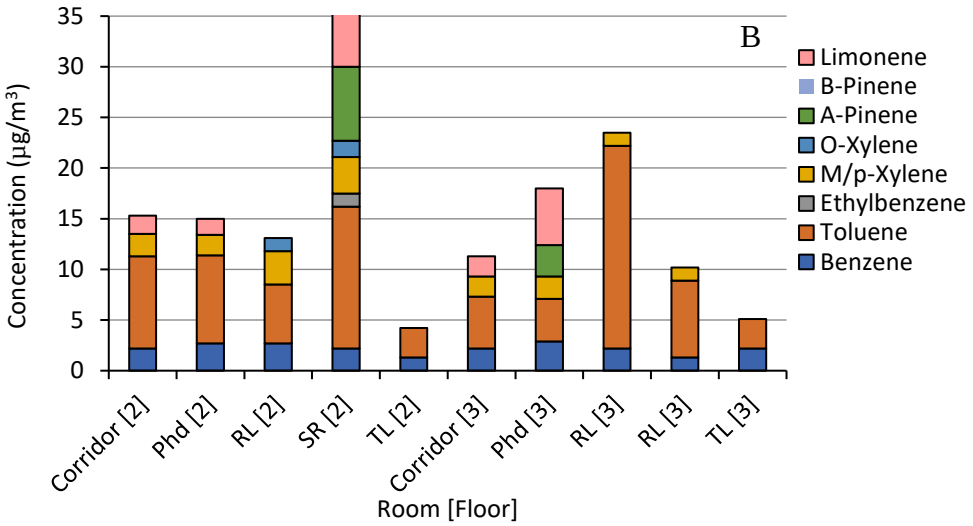


Figure 6.5: Concentrations of BTEX and mono-terpene compounds measured in different rooms within the Chemistry building. The locations of samples from Survey 1 (A) and 2 (B) are illustrated in Figure 6.1. A singular sample was taken from each location. Two samples are taken from RL2 and RL3 hence the duplication in records. Each concentration based on 1 "grab" sample taken from each location in November 2018 (20 minute grab sample at 500 ml/min) (A) and January 2019 (15 minute grab sample at 300 ml/min) (B).

Benzene and toluene were the most prevalent compounds in both surveys, which were found at the highest concentrations in the PhD [3] and RL [3] locations respectively. The Student Room (SR) (2nd Floor [2]) had the highest total VOC concentrations of all the locations investigated when making a summation of these eight fully quantified compounds, and highest concentrations of limonene (5.8 µg/m^3), α-pinene (7.3 µg/m^3), o-xylene (1.6 µg/m^3), m/p-xylene (3.6 µg/m^3), ethylbenzene (1.3 µg/m^3) and toluene (14 µg/m^3) (Figure 6.5).

Since the highest concentrations were measured within the Student Room (SR) adjoining the Teaching Laboratory (TL) possible transference out of the laboratory was investigated by taking concurrent samples throughout the day (over 5 hours; before, during and after activity) from this laboratory and the adjoining Student Room. Higher concentrations of BTEX and monoterpene compounds were found outside of the laboratory than inside the laboratory (Figure 6.6). However, this is largely attributed to

the presence of monoterpenes (2.9–6 µg/m^3) in these samples, which were absent from those taken from within the laboratory. These monoterpene compounds are widely used in cleaning products. Their absence in the laboratory highlights that the laboratory may have been cleaned less recently and/or the compounds have been removed through enhanced ventilation by local exhaust systems.

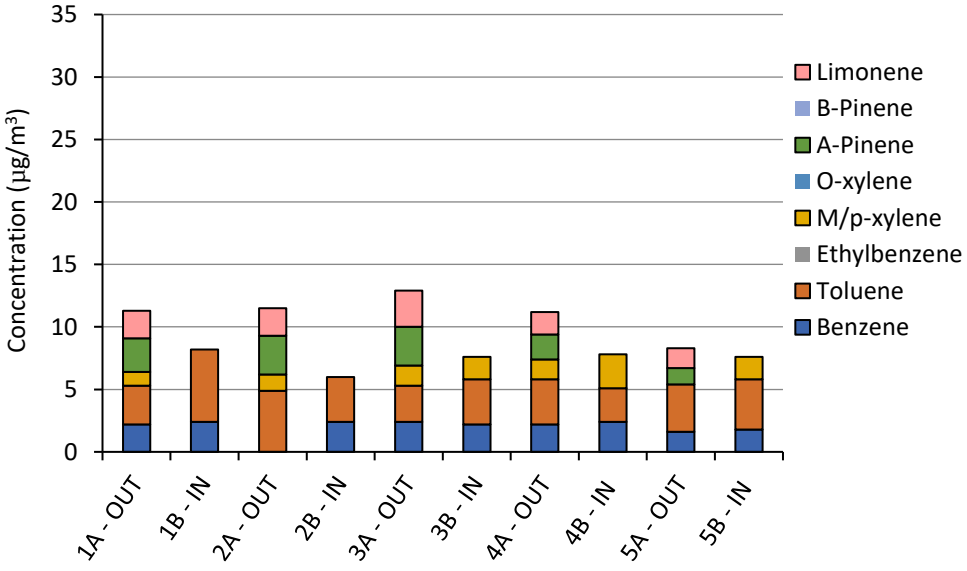


Figure 6.6: Concurrent samples throughout the day from inside and outside of teaching laboratory [B Floor] over course of several hours before, during and after known activities. Each concentration based on 1 15 minute "grab" sample (at a flow rate of 300 ml/min) taken in March 2019.

Using the semi-quantified methods on samples taken in the previous surveys, concentrations of the top 10 most prevalent compounds can be compared (Figure 6.7–6.9). In the first survey it was very unusual and unexpected to observe the very high levels of menthol (49,000 µg/m^3; 2400 µg/m^3) and glycerol (30,000 µg/m^3, 19,000 µg/m^3) in the PhD [3] and Synthetics Laboratory (RL) on C Floor [3]. Whilst it is postulated that this could have reflected an e-cigarette or vaping signature no visual observation was made of this at the time of sampling. It could also be that these chemicals were used in experimental work. These elevated concentrations could not be replicated through repeat sampling, suggesting the source was an isolated event. The

lowest VOC concentrations were observed in the Non-Synthetics Research Lab on B Floor, likely due to inactivity in this room during the sampling period.

As expected, higher VOC concentrations were found inside the laboratory than outside (Figure 6.8; RL vs Other Rooms). This finding contradicts the previous assertion that there is a large amount of passive transfer or transference from the laboratory to the surrounding or adjoining areas. The differing prevalence and concentration of VOCs between the laboratory and adjoining PhD Room on this occasion highlights that there is enhanced dilution in the laboratory due to local exhaust ventilation systems (fume hoods) that indeed reduces transference out of the laboratory. It can also be assumed that this was the reason for enhanced concentrations of BTEX and monoterpene concentrations outside the laboratory noted previously.

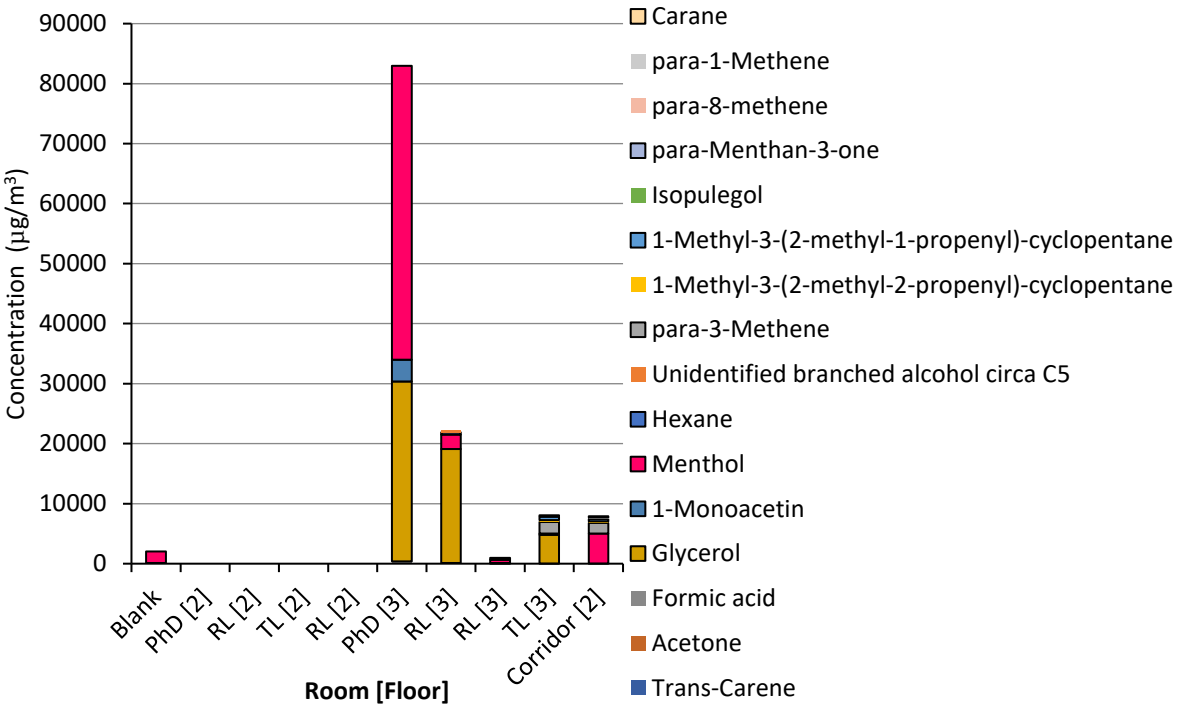


Figure 6.7: Semi-quantified (top 10 most prevalent compounds) from samples taken around the Chemistry building in Survey 1. Only compounds with concentrations above 100 µg/m^3 are shown, for further information see Supplementary Information 1.

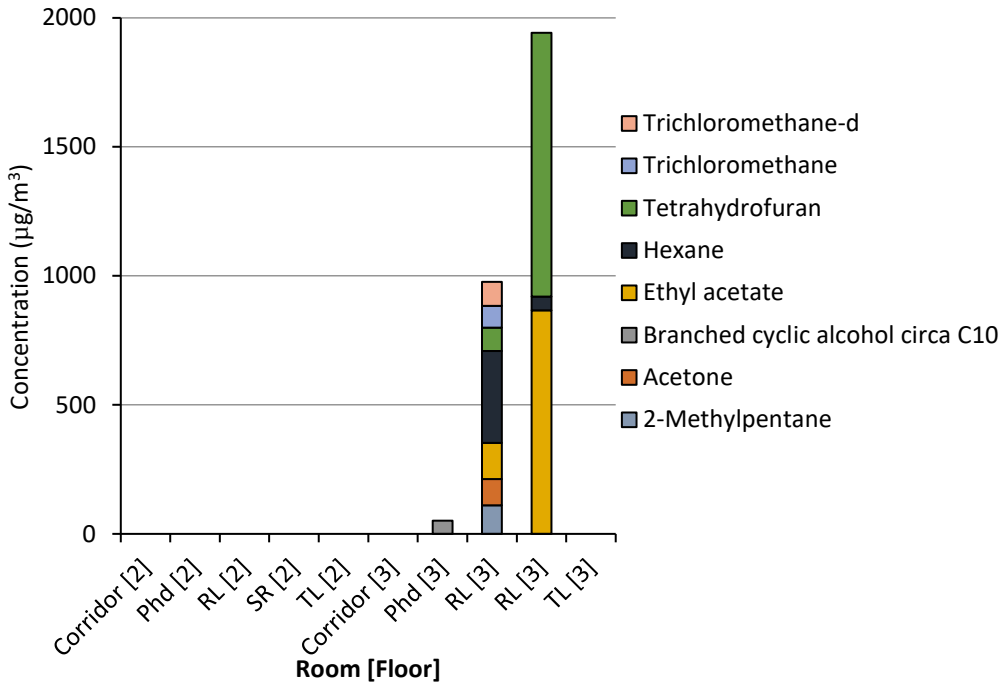


Figure 6.8: Most prevalent compounds for samples taken around the Chemistry building in Survey 2. Only compounds with concentrations above 50 µg/m^3 are shown, for further information see Supplementary Information 1.

The highest VOC concentrations were consistently observed in the Synthetics Research Lab (Floor C; RL [3]) which was to be expected due to the numerous known VOC emission sources. During sampling, multiple activities were being undertaken in the laboratory that would have contributed to this chemical footprint. Solvent and chemical use in the laboratories is thought to be responsible for higher VOC levels, as similarly found by Yurdakul et al. (2017). Chemical and solvent bottles exposed on shelves, disposal of small volumes of waste down the sink or bottles being left open for prolonged periods of time are all known to increase VOC levels (Valantidis and Vatistas, 2006). Even in fume hoods, heating of organic materials has been shown to increase VOC levels in previous studies (Yurdakul et al., 2017). It is difficult to attribute specific compounds to specific sources because there is no audit trail for the use of many compounds, and we are unaware of their use and location within the laboratory. We are therefore missing a level of granularity that would otherwise aid interpretation.

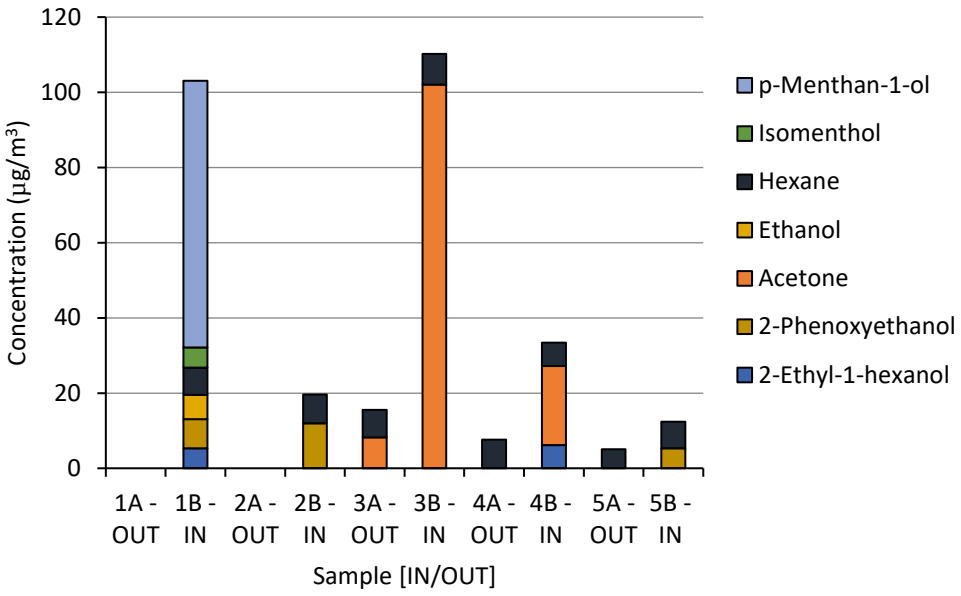


Figure 6.9: Most prevalent compounds from Survey 3 samples taken concurrently from inside and outside a laboratory over the course of a period of activity. Only compounds with concentrations above 5 µg/m^3 are shown, for further information see Supplementary Information 1.

The concentrations of VOCs observed in this study have been compared to national and international indoor air quality guidelines and occupational health exposure limits. No VOC levels exceed UK and EU occupational health exposure limits with the exception of benzene as the WHO guideline of "no safe limit" was exceeded in all indoor locations sampled. This no safe limit is established because of the carcinogenic nature of benzene. The concentrations of hazardous air pollutants measured in this study are similar to those observed in other university studies of air quality (Goodman et al., 2018; Godwin and Batterman, 2007; Zhang et al., 2006). Some studies only quantified a limited number of compounds, making comparisons difficult (Park et al., 2014; Goodman et al., 2018). Our initial extremely high levels of menthol and glycerol are not comparable with any other literature and these compounds are not in the top 10 most prevalent in all studies.

Zhong et al. (2017) observed low VOC concentrations in conventional schools and sustainable schools (mean < 5 μg/m^3) with the most prevalent compounds being the aromatics e.g., toluene, benzene, m/p-xylene and 1,2,4-trimethylbenzene (Zhong et al., 2017). Whilst similar compounds and concentrations were found initially during campus-wide sampling, much higher concentrations were observed during repeated sampling in the Chemistry building due to known strong emission sources. Concentrations recorded in this study were similar to those reported by Park et al. (2014) including for acetone and toluene. Benzene concentrations were lower than those reported by Park et al. (2014) but are not widely different; xylene concentrations were similar for both laboratories and corridors. It is postulated that the VOC compounds we identified could be attributed to similar sources identified previously, including off-gassing from building materials and household and cleaning products, and find high polluting activities that involve the storage and application of chemicals and solvents to be a significant source in laboratories (Bari et al., 2015; Rumchev et al., 2003; Valantidis and Vatistas 2006).

6.3.2.2. Physics Building

Owing to the recent (2016) refurbishments in the Physics building, not long before the sampling period (2018/2019), it is likely concentrations measured are a result of primary emission from building materials, products and furnishings. Building age has been shown to influence the concentration of carbonyl compounds. Molloy et al. (2012) and Langer et al. (2015) found that compared to older conventional dwellings, recently refurbished and energy efficient dwellings had increased concentrations of carbonyl compounds owing to increased ventilation and lower building related emissions in older dwellings. Higher VOC concentrations are also observed in new or renovated buildings

because of the application of many materials that are significant pollution sources, including water-based paints, adhesives and wood-based panels.

In terms of the spatial distribution, higher total and individual aldehyde concentrations were observed on C Floor [3] where was anecdotal evidence suggests there have been more recent refurbishments (Figure 6.10). In some circumstances, smaller rooms, or those with lower air change rates (those used infrequently or that remain closed/locked) have higher concentrations, with the exception of the atrium. This was to be expected as lower air change rates promote accumulation of indoor contaminants such as those emitted from building materials and furnishings. The low air change within C064 and C020 in particular are postulated to have an effect on pollutant concentrations.

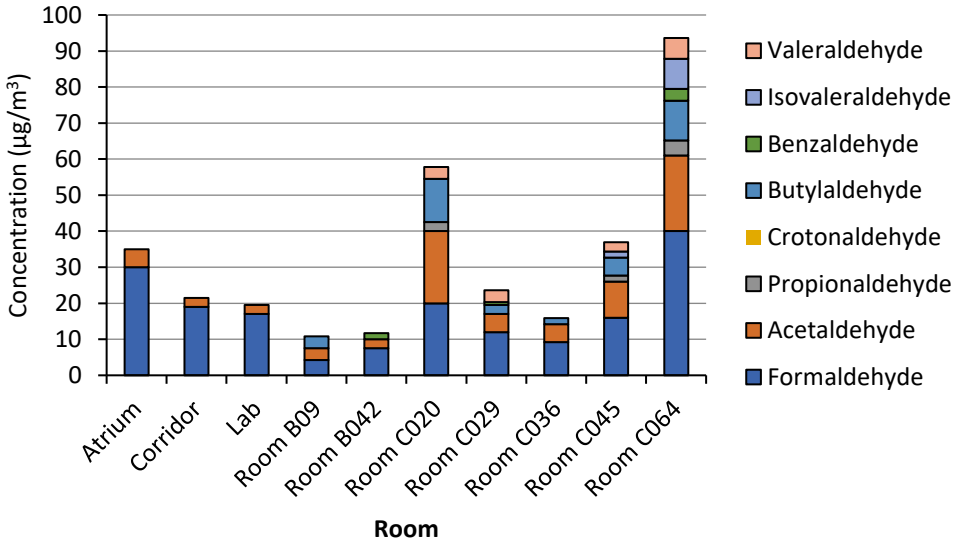


Figure 6.10: Quantification of eight aldehydes in each sample locality within the Physics building. Each concentration based on a single grab sample with a sampling period of 4 hours at a flow rate of 500 ml/min taken from June 2019.

The most prevalent aldehyde found was formaldehyde, with highest concentrations in Room C064 (40 µg/m^3) and the atrium (30 µg/m^3) (Figure 6.10). This was not unexpected since formaldehyde is ubiquitously found in the environment and a common component in many building materials and furnishings. Although the actual sources of

the formaldehyde could not be identified, most laboratories, classrooms and offices have plenty of structural materials (such as foam insulation and wood-based materials), furniture, interior finishing materials, and chemicals which can slowly emit formaldehyde (Valantidis and Vatistias, 2006; Kang et al., 2017; Destaillats et al., 2006; Singer et al., 2006; Kruza and Carslaw, 2019). Formaldehyde has also been associated with emissions from motor vehicle exhaust, (Kaden et al., 2010) but this is unlikely the case here due to the absence of vehicular traffic. Acetaldehyde is similarly prevalent in most samples, the highest concentrations in rooms C064 (21 µg/m^3) and C020 (20 µg/m^3). Its presence is similarly likely to be a consequence of recent refurbishments made throughout this building and indoor chemistry.

There is considerable guidance and advice on the use of materials such as timber/wood flooring which may contain formaldehyde in BREEAM assessment notes. However, it has not been possible to gain further information on the building and construction materials used in each building or area investigated in this study and therefore it is difficult to make any conclusions about their potential for formaldehyde contamination and difficult to assess the relationship between IAQ credits and formaldehyde sources. The lack of an audit trail collected at an institutional level for formaldehyde alongside other aldehyde sources makes interpretation difficult. An assumption is made that guidance was followed based on BREEAM assessment, and that contractors were working under best practice, but the poor records and difficulty in retrospectively trying to assess and identify sources makes it challenging.

This study found similar concentrations of formaldehyde to those found by Goodman et al. (2018) in meeting areas, classrooms and renovated offices, and restrooms and Shendell et al. (2004) in classrooms but Akal et al. (2015) identified higher

concentrations. Indoor formaldehyde concentrations ranged from 3.1 to 46.1 μg/m^3 and from 4 to 100 μg/m^3 in classrooms in Germany and France respectively and between 10.5 to 70 μg/m^3 in university offices and lecture theatres (Kaden et al., 2010). In the EXPOLIS study in Helsinki, similar levels of formaldehyde (15 μg/m^3) were found in the workplace (Kaden et al., 2010). These concentrations are within the range of those we observed from the Physics building.

6.4. Discussion and Critiques of Guidelines and BREEAM

6.4.1. Concentrations of Concern and IAQ Guidelines

Air quality guidelines are based on a review and interpretation of globally accumulated scientific evidence linking exposure to a selected pollutant with the health outcomes of that exposure (World Health Organisation, 2010). There are guidelines for some indoor pollutants, but these are not often enforced. Short and long-term indoor concentration guidelines are given, by the UK Government and the WHO (World Health Organisation, 2020). Long-term concentrations are not exceeded however there is no direct comparison between the time period of sampling and exposure periods for the guidelines.

The WHO has issued indoor air quality guideline values (World Health Organisation., 2010). A 30-minute guideline of 0.1 mg/m^3 of formaldehyde is been recommended to prevent sensory irritation in the general population. In our study we do not observe any formaldehyde concentrations above this limit, however there is a discrepancy between our sampling period and the WHO exposure time guideline recommendation. Benzene is a genotoxic carcinogen in humans and no safe level of exposure can be recommended. In the present study, the presence of any concentration of benzene can be considered unsafe due to the nature of this pollutant. However, there is no information on long term

concentrations. IAQ standards for important VOCs such as benzene and formaldehyde need to be developed for non-occupational exposure which often occurs over much longer time scales.

Short (30 minutes/1 hour) air quality guidelines have recently been proposed by the UK government for individual VOCs including formaldehyde, acetaldehyde, limonene and α-pinene (Public Health England, 2019). A comparison of concentrations from this study against these guideline values can be made. Short term concentrations of α-pinene (45,000 µg/m^3; 30 minutes), d-limonene (100 µg/m^3; 30 minutes), formaldehyde (100 µg/m^3; 30 minutes), acetaldehyde (1,420 µg/m^3; 1 hour) and toluene (15,000 µg/m^3; 8 hour) were not exceeded; however, there is a discrepancy between our sampling period (15 minutes) and the toxicological guidelines. Our sampling methodology did not account for the variability of concentrations over time and so extrapolation from a 15-minute sample is difficult. Wider implementation of these guidelines would be beneficial for assessing concentrations of concern in the context of toxicological information.

6.4.2. BREEAM Certification

In general, the chemical patterns or footprints are building dependant, with no clear relation to varying sustainability credentials. Similarly, building type (conventional vs high performance) did not appear to significantly influence VOC concentrations in studies by Zhong et al. (2017) or Verielle et al. (2016). The lack of an association between VOC concentrations and BREEAM certification could be related to the large diversity of buildings and associated characteristics but also to the process of BREEAM certification. Postulated reasons for this lack of relationship between BREEAM certification and VOC concentrations are discussed along with some clear thoughts

about how useful BREEAM guidance and air quality standards are for reflecting good IAQ and improving IAQ. Whilst this study is not necessarily providing new evidence around building performance in relation to IAQ, recommendations are made that would improve the assessment of IAQ in BREEAM accreditation in the post construction and post-occupancy stages of a building.

6.4.2.1. Is Current BREEAM Certification Fit-for-Purpose and a True Reflection of Good IAQ?

There are many useful attributes to BREEAM certification. It covers a vast array of topics on building sustainability and recognises and reflects the value in higher performing assets across the building environment lifecycle, from new construction to in-use and refurbishment. However, BREEAM is designed for performance buildings with a broad appreciation of IAQ that may not necessarily translate into something meaningful for building users. In other words, the value of an "Excellent" rating may be at best questionable and at worst misleading to building users. Many questions arise; is this the rating in the buildings current form? How is the building's sustainability and its IAQ changing over time? What does this mean for health and well-being, bearing in mind that there are no official IAQ standards to relate back to. The lack of an association between BREEAM certification and VOC levels has highlighted their divergence.

Credits are awarded to performance areas in each category that meet or exceed good practice (Bunz et al., 2006). These credits are weighted to take into account the importance of that particular issue in the overall environmental impact of a building (Bunz et al., 2006). There are only a few credits attributed to air quality and these are split between minimising sources of air pollution and ventilation (natural and provision of fume cupboards) (BREEAM, 2016). There are only four credits that relate to

minimising sources of air pollution, two of which relate to VOC emissions, one for the products used in buildings and one for post construction measurement (BREEAM, 2016). The former requires that all decorative paints and varnishes specified must meet certain criteria. The latter requires measurement of formaldehyde and total volatile organic compounds (TVOC). There are two standards that provide guideline values for these compounds. Formaldehyde has to be less than or equal to 100 $\mu g/m^3$. This is the same as the WHO air quality standard. TVOC concentrations have to be less than 300 $\mu g/m^3$. Where VOC and formaldehyde levels exceed these limits defined, the project team must confirm the measures that have or will be taken in accordance with the IAQ plan to reduce levels to within these limits (BREEAM, 2016). The guidance provided by BREEAM includes some specific information about "removal and/or dilution of sources" (BREEAM, 2016).

6.4.2.2. Critiques of BREEAM and Key Recommendations

It has been previously postulated that conventional and sustainable buildings do not show systematic differences in VOC levels and that there is no effect of building type discerned, due to the large diversity of university buildings and their building systems (Zhong et al., 2017). Buildings within a building type (similar age and airtightness) vary in terms of typology, HVAC system, furnishings, use, degree of crowding and other factors (Zhong et al., 2017). The characteristics of the building and the activities occurring within them contribute to their unique chemical footprint. A main finding from this study is that new and recently refurnished buildings and their building systems are diverse and not easily generalisable and separable from an IAQ perspective. We also see groups of VOCs are not very different i.e., building related and activity related VOCs.

Buildings can receive high BREEAM certification without consideration to good IAQ and without regard to VOC emission levels. Therefore, perhaps there would not be any expected relationship. This could explain the reason for the lack of association between VOC concentrations and BREEAM certification observed in this study. It could be that these credits do not fully reflect the importance of IAQ and thus do not reflect good IAQ. It could be argued that current credits are unsuitable for promoting better IAQ in green buildings and it is questionable as to whether BREEAM offers adequate incentives in the credit system for improving IAQ and pursuing these credits (Steinemann et al., 2017).

Questions have arisen regarding the usefulness and relevance of current BREEAM certification regarding these aspects. It is important to consider ways in which BREEAM assessment could be made more relevant for IAQ assessment without impacting other important IEQ parameters.

Recommendation 1: Integration of Continuous TVOC and Detailed Speciation

How useful a TVOC measurement might be in regards to BREEAM standards is a matter for debate. TVOC is used as a measure giving a possible indication of poor/good indoor air quality; furthermore, it is proposed as an indicator for the calculation of ventilation rate. However, TVOCs reveal little regarding the nature of the individual compounds, their concentrations and their possible toxicity to humans. It is clear that speciation is important due to the health effects of specific compounds, but these are currently not considered in BREEAM certification (with the exception of formaldehyde). This study found differing levels of a wide range of compounds attributed to numerous sources, but these data need to be combined with toxicological information to determine any risk. Benzene, a prevalent VOC in this study, is a

genotoxic carcinogen, where no safe level of exposure can be recommended, highlighting the importance of understanding concentrations of some specific compounds.

With the advent and rise in use of low-cost sensors it is now possible to achieve high temporal and spatially resolved measurements of TVOC concentrations to an accuracy comparable to reference instrumentation. It is possible that as sensor technology improves it will be possible to characterise (and understand) the TVOC signal. This could allow continuous TVOC monitoring throughout a building, which might tell us something about VOC exposure over time (as we investigated in this study) pre and during occupancy which BREEAM does not currently consider.

Understanding temporal patterns of VOCs in this way would give a starting point to isolate activity-based VOC signals from background VOCs. This could be complimented by tube sampling and detailed VOC speciation which was undertaken in this study. Grab samples currently used to assess IAQ credits in BREEAM only provide limited insights into sources of VOCs. Speciation and continuous TVOC monitoring with the advent of low-cost sensors could provide a better understanding of VOC patterns in space and time which could complement existing BREEAM approaches and in the longer-term lead to more meaningful BREEAM standards. This is an important recommendation from this study seeing as the current BREEAM guidance and air quality standards need to be improved for improving IAQ.

Recommendation 2: Inadequate Consideration of Activities and Location

To achieve BREEAM IAQ credits, measurements of TVOC and formaldehyde emissions are undertaken. However, assessment for BREEAM certification takes place in either the design or post-construction stage of a building. There is currently no

assessment of air quality in the post-occupancy stage. As we observe in this study there are significant differences and pollution signals between occupied and un-occupied monitoring periods especially in locations where strong polluting activities occur. Certain activities strongly influence pollutant concentrations within buildings. Activities within buildings, which occur independently of any certification, as seen in this study, play an important role in the prevalence and concentration of VOCs, and are not currently considered in BREEAM certification. This is a major limitation of BREEAM assessment. How people interact with buildings is an important determinant of IAQ and pollutant concentrations and it should be factored into the design of buildings for example, improvements in air handling.

It is therefore important to take measurements of pollutant concentrations in the post-occupancy stage to assess relationships between pollutant concentrations and activities (and building related emissions overtime). This is an important recommendation for BREEAM accreditation. A key recommendation would be that additional credits need to be awarded to various aspects of IAQ including the monitoring of emissions, particularly during polluting activities. BREEAM accreditation is awarded at the point a building was assessed, but it is not known how the building will change over time, and how pollutant concentrations will change as a result of changes as the building ages and different activities are undertaken. It would be beneficial to provide further quantitative assessment in the post-occupancy stage and then follow-up sampling of VOCs and IAQ in order for the building to maintain the BREEAM status further on. An expiry date for a BREEAM certification could be beneficial. The responsibility for post-build IAQ would lie with the occupants/renters or building managers, and monitoring could be undertaken by a team of trained professionals within BREEAM.

The locations of measurements around and within a building need consideration. Whilst it is important to measure IAQ in general facilities and areas, IAQ also needs be investigated in specialised (e.g., laboratory) areas with high polluting activities. Temporal and spatial signatures of the source can be elucidated, enabling better source control.

Recommendation 3: More Credits Awarded For Volatile Organic Compounds

Whilst concentrations in this study remained within those required by BREEAM VOC emission credits and occupational health exposure limits, this study was highly time sensitive, and it could be said that BREEAM and other schemes need to place more emphasis on VOC emissions as a subset of IAQ due to the health effects of enhanced VOC levels. This is especially important as "green" does not guarantee good IAQ. Whilst green and sustainable buildings may promote energy efficiency and sustainability, they do not necessarily promote the health and wellbeing of occupants through better air quality (Steinemann et al., 2017). Certain green practices and products can impair IAQ since green products do not need to disclose all ingredients, which can include hazardous air pollutants such as waste-based or recycled materials and they may lack verification and monitoring of their emissions (Steinemann et al., 2017).

Recommendation 4: Greater Emphasis on Source Control

This paper shows the challenge of source identification and therefore why control is so difficult. Source control is relatively straightforward in cases where sources are obvious and easily replaceable but difficult where the primary sources are construction materials and the building fabric as in this study. Nonetheless source control is important, often noted as the first line of defence for removing emissions. The focus on ventilation in

BREEAM certification may overlook opportunities for source control (Steinemann et al., 2017).

6.5. Limitations

There are several limitations associated with the methodology, in particular the sampling design and analytical method chosen. The duration of sampling required to collect sufficient volumes to meet MDLs differed from duration for health-based guidelines preventing direct comparisons in some situations. With regards to using the GC-MS for quantification of VOCs, oxidised compounds and carbon compounds below C_4 including ethanol were not measured. Due to the numerous possible VOCs that could have been observed, semi-quantitative methods were chosen alongside full quantification to focus on identification of compounds. Semi-quantitative methods are less accurate than full quantification with external standards. Further research could focus on full quantification of the compounds identified in this study. A further limitation of this present study was a difficulty in retrospectively associating sources of VOCs to VOC concentrations, particularly where there may be simultaneous presence of multiple potential primary and secondary emission sources. Whilst frequently used chemicals in each laboratory were investigated it was not possible to find out exact amounts of chemical used during sampling and how they were used as well as laboratory conditions therefore source characterisation was difficult.

6.6. Conclusion

The aim of this study was to assess the concentration of VOCs in sustainably accredited buildings, to evaluate VOC sources and assess the extent to which BREEAM accreditation is a reflection of good IAQ, and whether IAQ credits are fit for assessing building sustainability and promoting health and well-being related outcomes.

In conclusion university indoor environments can be important sources of pollutants. However, in this study overall VOC concentrations were low, and in many cases not higher than the limit of detection. This study postulates VOC emission sources though it is difficult to provide a definitive association between source and concentration due to the vast number of potential emission sources. The majority of VOCs were thought to be attributed to activities within and throughout the buildings. A lack of audit trail on materials used and ongoing activities collected at an institutional level hindered this investigation. The VOC concentrations within buildings housing laboratories were much higher than concentrations within non-laboratory buildings due to the large amounts of chemicals and solvents used within these buildings. It was interesting to observe higher concentrations of BTEX and monoterpene concentrations in PhD Write Up or Student Rooms adjoining but closed off from laboratory environments (with known VOC emission sources). It was originally postulated this could be attributed to passive transfer out of the laboratory. Further sampling suggested enhanced dilution of building and cleaning product contaminant emissions in the laboratory due to local exhaust systems was responsible. Formaldehyde and acetaldehyde were the most prevalent aldehyde compounds seen, which are ubiquitous in the indoor environment. Tentative conclusions are made between the spatial distribution of these compounds and recent refurbishment in the Physics building, which experienced the highest aldehyde concentrations around campus.

There appeared to be no association between BREEAM certification and VOC levels, and no significant differences noted in the chemical footprint between recently built, energy efficient university buildings, recently refurbished BREEAM certified buildings and conventional buildings. The lack of discernible difference in VOC concentrations between buildings of various sustainable accreditations and those without was not

necessarily surprising and could be attributed to the large diversity of buildings and their systems. We conclude that green buildings with sustainability credentials do not necessarily have better IAQ than conventional buildings and this is largely in response to the activities undertaken within individual buildings. We postulate that the lack of significant IAQ differences between buildings owes much to the way in which BREEAM accreditation is constructed and could be a response to the lack of credits awarded for IAQ and VOC emission levels. Activities within buildings, which occur independently of any certification, as seen in this study, play an important role in the prevalence and concentration of VOCs, and are not currently considered in BREEAM assessment. More consideration could be given to activities within buildings, and not solely the building fabric, in the certification process. This study makes recommendations as to how BREEAM certification can widely encompass VOC emission levels based on observations made. A framework of integrated continuous TVOC monitoring and detailed speciation of specific VOCs in both the post-construction and post-occupancy stages of a building is suggested to better understand spatial and temporal VOC patterns and differentiate between fabric-related and activity related emissions to give a better measure of IAQ.

6.6.1. Recommendations

A limitation of the present study was the difficulty in retrospectively associating sources of VOCs to VOC concentrations, particularly where there may be simultaneous presence of multiple potential primary emission sources, from the building or occupants, and potential secondary source (Verielle et al., 2016) or where there may be no obvious emission sources. This made source apportionment and characterisation difficult. If information about experimental conditions, physiochemical properties of the chemicals,

and by-products caused by chemical reactions could be obtained, the source of target VOC compounds could be identified more accurately. This could be a focus of future work. Reliable conclusions could not be taken from this study based on statistical analysis regarding VOC levels across different building types (conventional vs sustainable buildings) and within buildings due to limited sample size. To be fully comprehensive and generalisable, a larger set of buildings should be studied and more homogeneity in season and building attributes (energy consumption or ventilation control schemes) should be incorporated (Verrielle et al., 2016).

Finally, a key recommendation is to test the methods prescribed here that target better characterisation of VOCs and IAQ for BREEAM accreditation through a combination of continuously monitoring TVOC signals and taking grab samples for detailed VOC speciation in the post-occupancy stage of a building. Long term VOC exposure was not within the scope of this present investigation but through combining these two techniques we could better understand long term exposure patterns. Capturing long-term exposure and toxicological information would enable better understanding of activity related influences and primary and secondary building related emissions that may last over longer periods (Sundell, 2004; Prasaukas et al., 2016).

7.0. Summary and Recommendations

This thesis encompassed a broad range of research that focussed on and addressed research gaps within the field of indoor air quality (IAQ). Prior to undertaking the research, a literature review was conducted to summarise existing knowledge and identify knowledge gaps that were assessed in subsequent chapters. Through this review, the need for further research into source characterisation was identified, along with research into trade-offs between IAQ and energy efficiency.

The overall aim of this thesis was to evaluate the sources of indoor air pollution (with a particular focus on ultrafine particles and volatile organic compounds) and the controls on IAQ within residential and educational micro-environments. It also aimed to evaluate the potential consequences of improving IAQ through examining the dichotomy between good IAQ and energy efficiency. To achieve these overall aims, the research addressed two main objectives. First, to better understand the spatio-temporal patterns of indoor air pollution from typical household activities in both real-world and controlled residential micro-environments. Second, to better understand the prevalence and concentration of volatile organic compounds (VOCs) across a University campus and establish whether any relationship existed between BREEAM building accreditation and VOC levels. A further objective was to evaluate the practicalities of using V2000 air quality monitoring units developed by NAQTS (a start-up company who partially funded this research). These units show potential to bridge gaps in IAQ monitoring knowledge and raise public awareness of IAQ issues in public and private settings.

This thesis adopted a range of different research methods including a critical evaluation of existing literature into themes including source characterisation, IAQ monitoring

technologies and exposure mitigation. An extensive field campaign was undertaken across 8 residences in the NW of England using NAQTS V1000/V2000 monitoring units. This was followed by a more focussed study within a controlled residential environment using the same sampling protocol. Finally, VOCs and carbonyl compounds were sampled within 10 buildings across a university campus, with further localised sampling within hotspots of activity, in accordance with regulatory methods and standards.

This chapter summarises the research that has been carried out in this thesis and discusses key findings and implications. Each of the experimental chapters (Chapter 4, Chapter 5, and Chapter 6) contains a unique discussion. This chapter summarises the main research outcomes, considers overarching themes and the implications of the research findings. Suggestions for further research are also presented, in response to, these findings.

7.1. Summary of Research Outcomes

Chapter 1 provided an introduction and context and statement of the problem. It also gave a brief overview of the knowledge gaps and a summary of the main research aims. This was expounded in Chapter 2 which provided a detailed literature review that identified key knowledge gaps addressed in this research. Chapter 3 provides a technical specification of the instrumentation used to evaluate indoor air quality in residential micro-environments in Chapters 4 and 5 of this thesis.

Chapter 4 presented the results of a pilot study that aimed to evaluate spatio-temporal patterns of indoor air pollution associated with typical episodic household cooking activities. It assessed the controls on particle number concentrations (PNC) including ventilation and housing layout. It is important to understand potential exposure to

particles generated by cooking to understand the health implications. The objectives were to assess the influence of episodic cooking activities on particle number concentrations, taking high time-resolved measurements for 7 non-consecutive days in 8 homes in NW England, and to evaluate the influences of natural and mechanical ventilation, and housing structure on the decay of ultrafine particles.

In this study we observed that episodic cooking activities generated high peaks in PNC quickly after the onset of cooking activities which decayed at a rate strongly dependent on the air exchange rate. We computed descriptive and analytical statistics across all source categories and ventilation scenarios and found that significant differences in PNC could be attributed to differences in ventilation. Ultrafine particle emissions resulting from the three cooking methods suggest that cooking in a house with inadequate ventilation could lead to indoor concentrations that exceed those outside, and that could negatively affect the health of occupants. Natural ventilation was generally seen to be the most effective means of reducing PNCs associated with discrete cooking activities.

We conclude that a single V2000 monitor can gain a representative measure of PNC in a single room in a domestic setting. In multi-room scenarios, where we simultaneously monitor PNC response to cooking in multiple locations, we find cooking-generated PNC are not only high in the kitchen, but in other locations around the house, which we attribute to housing layout. Potential health consequences may be underestimated if whole house dynamics are not considered.

This study provided insights into controlling influences on cooking emissions and the influence of housing layout on internal airflow. In this pilot study only a limited number of properties were considered due to time constraints and sensitivities involved in

monitoring in real-world environments. This limited our ability to statistically assess differences in PNC between different types of housing. In the future, this study could be repeated with a larger and more diverse range of properties. Whilst natural ventilation was seen to be the most effective strategy for reducing exposure to cooking-generated particulates, the energy consequences of this are potentially high, particularly in the heating season.

Chapter 5 explored the energy consequences of exposure mitigation to particles generated by typical household cooking events in a controlled residential environment. V2000 units were deployed within The Salford Energy House to examine PNC response to discrete cooking activities under different ventilation scenarios and assess the potential energy penalties associated with these different ventilation strategies.

In the previous chapter we made a recommendation to understand the practical implications of using ventilation including the energy consequences. The novelty of this chapter lies in its attempt to better understand the energy consequences of ventilation and ventilation heat loss to better manage sometimes conflicting objectives of promoting healthy IAQ and maintaining energy efficiency. Independent measurements of PNC and ventilation heat loss were conducted under different ventilation scenarios and energy penalties were calculated based on the amount of heat lost to the external environment. Regardless of the type, we highlight the importance of ventilation for improved IAQ.

Generally, energy penalties from either natural or mechanical ventilation were low on the order of a single experiment. Energy generated by cooking processes led to an energy surplus in some instances when opening windows or operating extract ventilation for short periods of up to 10 minutes. We identified an optimal period of up

to 20 minutes of window opening when ventilation delivers an improvement to IAQ with no significant energy consequences. Beyond this, we observed more significant heat losses. The energy used by an extractor fan is also negligible and energy penalties are much lower, even given consideration to both space heating consumption and that used to operate the extract fan, than natural ventilation. Intermittent mechanical ventilation is therefore seen to be an important mechanism for improving IAQ without incurring energy penalties. Whilst we experience a negligible energy penalty for the period of operation we used (30-minutes) we suggest an optimal period of 20 minutes to better balance the dual objectives of good IAQ and energy efficiency.

We conclude that the energy penalty associated with natural or mechanical ventilation is negligible in the specific case of Salford Energy House, but acknowledge that in the real-world, these penalties (particularly for natural ventilation) could be significant given the greater pressure differential between internal and external environments. This study is applicable to a mid-terrace typical 1940s build in the UK and therefore does not extend to other dwellings. We cannot make any generalised assumptions or approximations for other housing types and this could be the focus of future research. There were limitations to using the EH as a testing facility, with regards to its operation compared to the real world. Driving forces operate differently in the EH, with respect to natural ventilation and pressure and temperature differentials which may have resulted in an under-estimation of energy penalties. Despite these shortcomings, this chapter helped to develop a methodology for promoting a balanced approach to building management that prioritises IAQ and energy efficiency. This balanced approach will be particularly important in the post-COVID world where the built environment and ventilation is seen as vital in minimising community transmission.

Chapter 6 investigated the prevalence and concentration of VOCs, including carbonyls, within a variety of indoor environments at a medium-sized UK University campus using a sieve mapping approach. The study aimed to evaluate sources of VOCs and the relevance of IAQ credits in BREEAM accreditation to determine whether the current accreditation approach was fit-for-purpose. The objectives were to determine the concentration of VOCs for selected buildings around a university campus (sustainably accredited BREEAM buildings and otherwise), to assess the association between VOCs and BREEAM standards, and assess the relevance and validity of IAQ credits in BREEAM standards.

University buildings can be important sources of air pollutants. However, low VOC concentrations below stated guideline values and BREEAM limits were observed across all buildings. High ventilation rates could be responsible. Buildings housing laboratories generally experienced higher VOC concentrations, particularly the Chemistry building, where synthetic chemicals and solvents were being used. Relatively higher BTEX and monoterpene concentrations were also measured in locations adjacent to laboratories in the Chemistry building, suggesting enhanced dilution of building and cleaning-related contaminants by local laboratory exhaust systems rather than passive transfer from the laboratory. Potential VOC emission sources were identified, although the lack of audit trail on materials used and ongoing activities collected at an institutional level hindered this investigation.

This study provided insights into the prevalence and concentration of VOCs in a multi-functional building environment and a solid foundation for further work quantifying VOCs within indoor environments. There appeared to be no association between BREEAM certification and VOC levels, and no significant differences noted in the

chemical footprint between recently built, energy efficient university buildings, recently refurbished BREEAM certified buildings and conventional buildings. The lack of association between a building's chemical footprint (VOC concentrations) and BREEAM certification leads us to question the value of IAQ credits in BREEAM standards. The novelty of this research lies in the suggestions made to develop the IAQ credits to provide a more relevant assessment of IAQ within buildings. Activities within buildings, which occur independently of any certification, play an important role in the prevalence and concentration of VOCs, and are not currently considered in BREEAM assessment. More consideration could be given to activities within buildings, and not solely the building fabric, in the certification process. While building accreditation schemes are principally concerned with building related emissions and minimising the ingress of outdoor air pollutants, this thesis suggests that to divorce the actions of people inside a building with the structure itself is short-sighted, particularly with regards to VOCs measurements. To facilitate activity-based assessments of IAQ, this study also recommends a new approach to quantifying VOCs that combines simultaneous temporal measurements with sensitive TVOC detectors alongside detailed speciation with reference instrumentation over longer time periods. This has been recognised in the WELL Standard, an international assessment method that encourages healthy choices and lifestyles as well promoting high standard of air quality (AECOM, 2020).

This PhD was part funded by National Air Quality Testing Services Ltd, and their involvement has been important throughout. Whilst the first two analytical chapters (Chapter 4, and Chapter 5) are clearly linked and highly reliant on the V1000/V2000 units, this chapter uses more conventional approaches to studying this aspect of IAQ. However, our recommendations were in part influenced by the potential capabilities of

these V2000 units which could be used to measure TVOC in real-time and collect thermal desorption samples of air for subsequent speciation through GC-MS.

7.2. Overarching Themes and Implications of Research

In the literature review we identified several areas for further research that focused upon understanding the causes, controls, and consequences of managing indoor air quality and energy efficiency dichotomies. Our evaluation of the research shows VOCs and particulates can be abundant indoors, suggesting a need to better understand emission sources and reduction strategies of these pollutants. However, improving IAQ should not come at the expense of energy efficiency, hence the importance of understanding the causes of poor IAQ and the ways in which it may be improved without incurring significant energy penalties.

This research utilised novel technology (V2000) that encompassed a variety of low-cost sensor technologies and regulation grade equipment for the monitoring of indoor air pollutants at high spatio-temporal resolution. We characterised sources and identified primary indoor air pollutants in residential and university micro-environments. We demonstrated that we could measure key indoor air pollutants to a reasonable degree of accuracy for multiple locations in an affordable manner. Importantly, we provided an understanding of the spatio-temporal evolution of air pollution episodes in properties of varying age and characteristics. We have also demonstrated how air pollution levels can be influenced by both local ventilation decisions (none, natural, mechanical) in the kitchen and air flows around a house (integration of Chapters 4 and 5). We have also begun to understand the costs of mitigation (the “energy penalty”), putting a financial and environmental value on decisions made to improve IAQ. As such, we begin to

understand ways in which we can harmoniously achieve the conflicting objectives of good IAQ and energy efficiency.

We can see how better information on IAQ can inform behaviour in both residential and non-residential environments, for both activity-related (e.g., cooking, cleaning, chemical-use) and building-related sources of pollution. We can also see how better and more easily accessible information on IAQ can better inform policy decisions at an institutional level and above. We consider it important to have IAQ information at a building level given the differences we observe between and within buildings, although we acknowledge the potential impracticalities of delivering this. It has been noted that significant differences exist between buildings based on age of construction, and that modern buildings are generally more airtight, potentially increasing air pollution levels in the absence of adequate ventilation. IAQ standards are emerging, but more work needs to be done in this area (BRE, 2019). These standards are important to assess IAQ in terms of negative health and well-being consequences. As more information becomes available, IAQ standards will be easier to identify, implement and regulate.

We can see how consideration of IAQ is important when designing new buildings, or retrofitting old ones, whether Passivhaus, BREEAM-rated or otherwise. Our research presents evidence to suggest that green buildings do not necessarily guarantee better IAQ. We can see that there are challenges for the building sector, for suppliers as well as developers, and that current green credentials may not be sufficient. We highlight the need for better integration of temporal and spatial VOC signatures when undertaking BREEAM assessments or other building assessments.

We know IAQ can be a problem and have demonstrated some of its characteristics and ways in which it can be controlled. We have shown that indoor air quality monitoring

can lead to better understanding of causes, controls, and consequences which in turn may lead to behavioural change (e.g., in domestic setting) and perhaps sectoral change in building construction and maintenance. This study has been important for raising awareness of IAQ and IAQ problems.

This thesis has also highlighted the importance of reducing domestic energy use to meet ambitious climate change targets. This research has shown how policy decisions should be sensitive to IAQ issues in addition to energy savings such that we can maintain healthy indoor environments in an energy efficient manner.

7.3. Opportunities for Further Research

The work presented in this thesis addresses some of the current research gaps in IAQ. There has been a notable increase in the number of publications on various aspects of IAQ in recent years, highlighting the rising importance of this emerging research area. Poor air quality is the largest environmental risk to public health in the UK, according to DEFRA's Clean Air Strategy (UK Government, 2019). However, this strategy is focussed very much on the external environment. There are many benefits to improving and maintaining good IAQ which include health and well-being outcomes. However, many important research challenges remain.

It is first and foremost of great importance that we promote awareness of IAQ and IAQ issues more generally, through widespread and affordable indoor air quality monitoring. Companies such as NAQTS have made significant progress in air quality consulting and wider air quality monitoring. They have recently been doing field campaigns in schools to understand the relationship between air pollutants and exposures. There are plans to lease monitors to clients for fixed periods of time to give them a better understanding of air quality issues. Creating or building an online database for such a

client base could lead to a better understanding of IAQ across the whole building sector, leading to performance ratings relative to sector norms for different types of buildings. This need has been recognised by UK bodies with much more emphasis on addressing air quality challenges at the indoor/outdoor interface of late. For example, as recently as July 2020, UK Research and Innovation has awarded £3 million to support multidisciplinary research networks to tackle major air quality challenges within both indoor and outdoor spaces including home, school, work, and public transport environments. The Small Business Research Initiative, delivered by Innovate UK also provides opportunities to innovate through emerging technologies (UK Research and Innovation, 2020).

The evaluation of PNC response to typical cooking activities (Chapter 4) revealed that the spatial pattern of the response is not well understood across houses of different characteristics. Further research could characterise household sources in a greater number of residences with varying characteristics, using a similar approach to the one adopted in Chapter 4. Understanding and having a means of comparative assessment (through a database perhaps) spatial patterns of particulates generated from cooking and other household activities is important as an activity in one room (e.g., kitchen) can impact on the IAQ in other rooms (e.g., living room, bedroom) and has potential exposure implications as residents may unwittingly spend long periods of time (e.g., overnight) in rooms with poor IAQ (e.g., bedroom). Modelling could also have a role to play here, for example, computational fluid dynamics models could be used to simulate air flows and pollutant concentrations in complex indoor environments.

We made some interesting observations in Chapter 5 and wider monitoring in similar controlled circumstances and also in real-world settings (though this comes with its

challenges) could enable more generalisations and approximations of the energy penalties and consequences of air quality exposure mitigation to be made for the wider building stock. During the lifetime of this PhD we were approached by Passivhaus to conduct IAQ measurements in their low energy houses (Passivhaus, n.d.). With strong airtightness requirements and better temperature control we may expect to see lower energy penalties than in conventional houses, though air pollution from human activities or building fabric and furnishings may still remain an issue. We would then also have to consider we may get further enhanced energy surpluses from cooking activities, and that we would then need to use ventilation for the purpose of thermal comfort.

In Chapter 6 we saw a need for a more integrated temporal VOC monitoring (with low-cost sensor technologies) and speciation (with GC-MS or similar technology) to assess VOC exposure in buildings and assess BREEAM certification. It is important to differentiate between fabric (building) related and activity related VOCs. The temporal VOC signal used in conjunction with activity diaries (and lack of activity records) is key to differentiating activity-related VOC spikes from other longer-term building fabric-related signals. We could not easily do this with our grab samples because of the time integration issue and lack of audit trails, but this could be a focus for future research.

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9.0. Technical Evaluation

This chapter discusses the case studies that are referred to in Chapter 3 that evaluate the use and practicalities of the NAQTS V1000/V2000 monitoring units that inform their use in the thesis.

Case Study 1: Student Accommodation (May 2017)

This case study was carried out on the 3rd May 2017 in a Lancaster University managed residence. The aim of this case study was to become familiar with the operation of the V1000 units in a real-world environment and to examine sensor response to occupancy and "light" cooking activities. As we can see from Figure 9.1 there are clear CO_2 spikes in response to occupancy in the bedroom (enhancements of 100–200 ppm) when in close proximity to the sensor. We can also clearly observe peak responses in PNC (Figure 9.2) from light cooking activities (toasting) in the kitchen followed by a significant reduction or decay owing to increased air exchange (window opening) after these events. We observed corresponding increases in CO_2 and CO in response to cooking activities and to a lesser extent, occupancy in the kitchen (Figure 9.3).

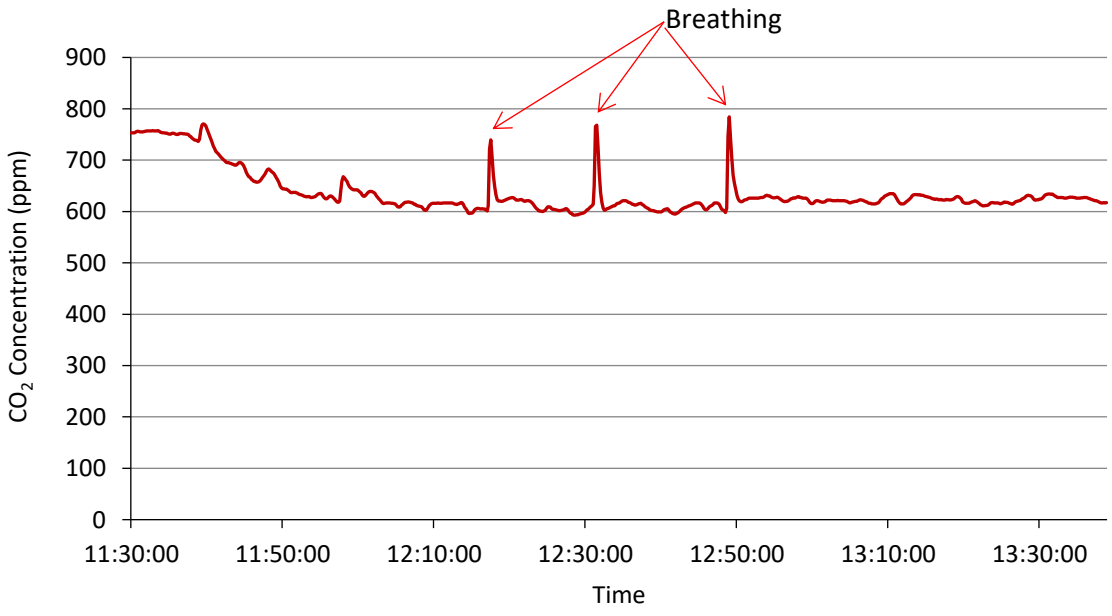


Figure 9.1: Temporal fluctuations in CO_2 concentration in response to occupancy in a student bedroom over several hours, as reported by the V1000 sensors.

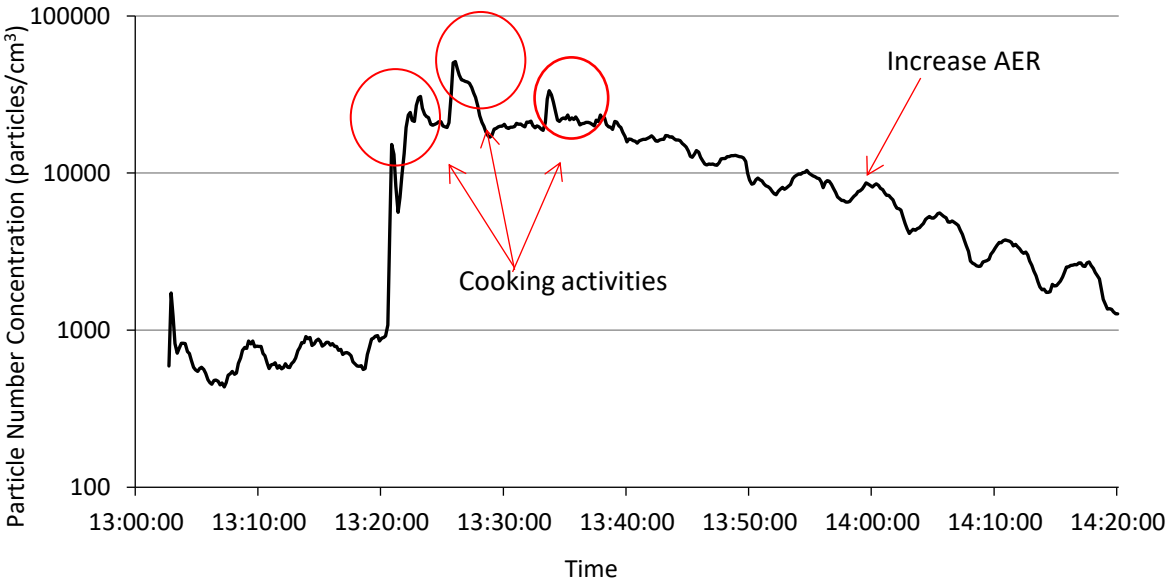


Figure 9.2: Temporal particle number concentration (PNC) response to light cooking activities (toasting activity) in a student residence kitchen over approximately an hour. Timing of cooking activities indicated as well as point of increased AER following toasting.

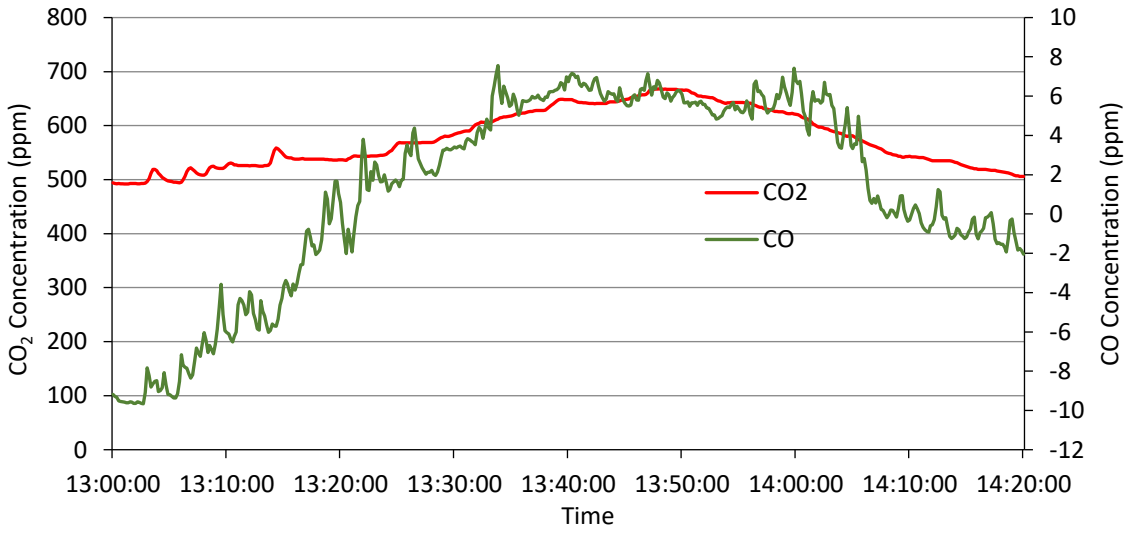


Figure 9.3: Temporal CO_2 and CO response, corresponding to PNC response previously shown (Figure 9.2), to light cooking activities and occupancy in a student residence kitchen over the period of just over an hour.

In this case study we noted the importance of regularly checking the reporting of the CO_2 and PN data online, as well as ensuring there were no problems with connectivity.

We also saw the importance of regularly ensuring fluid levels (IPA) were sufficient for the operation of the CPC.

Case Study 2: NAQTS Office (July 2017)

From the 17th July onwards a V1000 unit was placed in the NAQTS Office in the Gordon Manley Building in Lancaster Environment Centre (LEC) to monitor indoor air quality and measure pollutant concentrations. This case study was undertaken to look at sensor responses in a relatively pristine environment, free of known strong contaminant sources. This study was undertaken over several weeks to look at long-term trends in air quality data obtained by the V1000 units. Data were obtained at a resolution of 10 seconds. This report summarises results from the 17th–31st July 2017.

Carbon Dioxide

Carbon dioxide (CO_2) concentration is related to occupancy as expected. During periods of non-occupancy CO_2 concentrations remain at background concentrations, which are about 400 ppm (Figure 9.4). During periods of occupancy, CO_2 concentrations vary between 500 and 1900 ppm depending on occupancy level and occupant behaviour. Occupancy level significantly effects maximum CO_2 concentration for a given period. Once more than one person is in the office, there is a heightened CO_2 concentration as expected. It is the increase per person that is key not the overall concentration. Considering this CO_2 concentration appears to double in response to a doubling in occupancy level with the subtraction of background concentration. Occupant behaviour, including opening of windows (enhanced ventilation), affects CO_2 concentrations owing to its control (and effect) on CO_2 decay. Once closed, CO_2 concentrations can reach as high as 900 ppm and 1900 ppm for one and two persons in the office,

respectively. When windows are opened we see faster CO_2 decay attributed to dispersion and air exchange with external air with lower CO_2 levels.

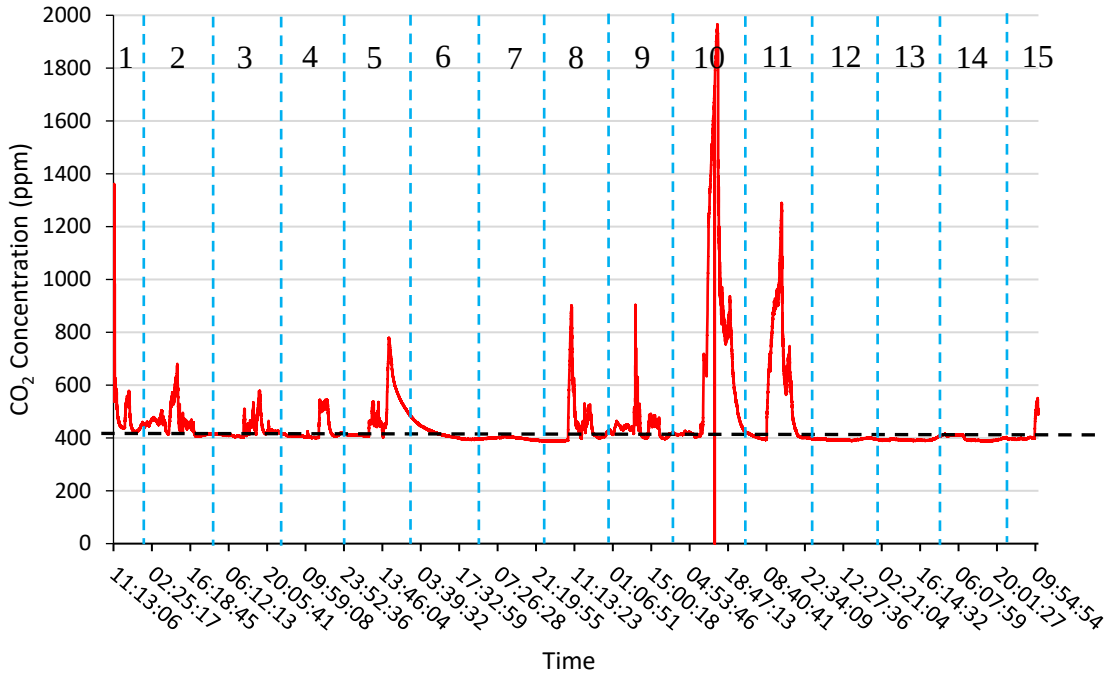


Figure 9.4: Carbon Dioxide (CO_2) concentration provided by NAQTS V1000 units over the measurement period (17th July at 11:13 to 31st July at 11:00am). Data is divided according to days of monitoring and these are labelled. Background CO_2 concentration highlighted at 400ppm.

Air exchange rate (AER) was calculated on several days (20th, 21st, 26th, and 27th) to establish an AER for the office and evaluate how it is affected by occupancy and occupant behaviour. We define air exchange rate or air changes per hour (which we abbreviate to AER) as a measure of the air volume added to or removed from a space (room or house). We calculate AER as a function of the logarithmic decay of CO_2 and subtracting the background value. AER is lowest on the 21st July (0.07 air changes per hour) (Figure 9.5A). Following occupancy on the 21st July the windows were closed so the CO_2 attains a higher level, attributed to occupancy, and thus we assume takes longer to decay. AER equals around 0.5 air changes per hour on the 20th (Figure 9.5B) and 27th July (Figure 9.5D) when the windows were left open following periods of occupancy,

permitting rapid CO_2 decay from the office (due to enhanced ventilation). AER reaches 0.2 air changes per hour on the 26th July as although the windows were left open following occupancy, we have significant inputs and concentrations of CO_2 which then decline from a much higher initial value (Figure 9.5C). Although not considered here, external wind speed would also be an influencing factor, controlling pressure differentials and hence air exchange between the indoors and outdoors. We conclude that we see higher AER rates during periods of enhanced ventilation (via window opening), likely attributed to greater pressure differentials driving the exchange.

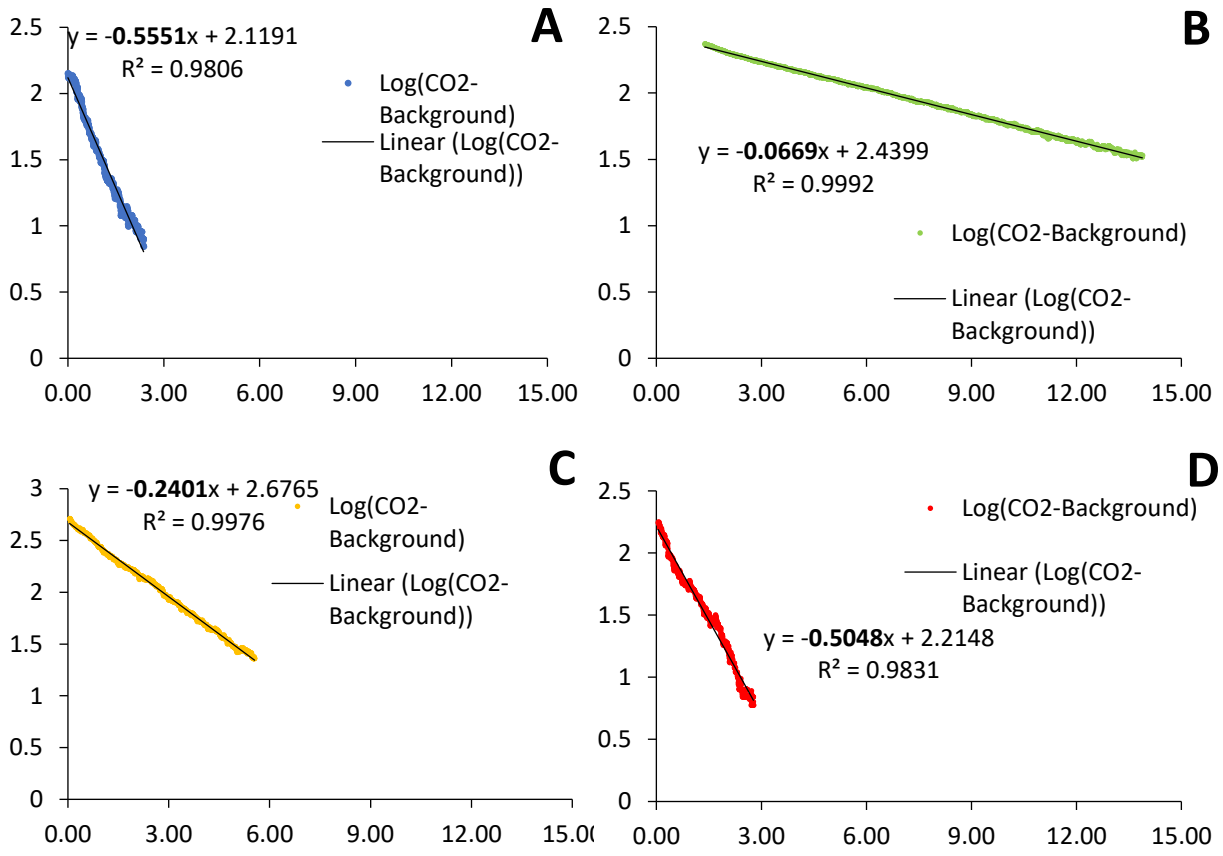


Figure 9.5: Air Exchange Rates (AER) at various periods throughout the measurement period, (A) 20th July; (B) 21st July; (C) 26th July; (D) 27th July calculated using CO_2 measurements and CO_2 decay after occupancy.

Particle Number Concentrations

Particle Number Concentrations (PNC) illustrate notable fluctuations (Figure 9.6). Very large, randomised peaks throughout the measurement period may be due to equipment error (in part random noise, and in part due to the technical errors) and can be removed from the time series by filtering data on diagnostic error flags (for example an inconsistent dilution ratio). Generally, PNC were below 10,000 particles/cm^3, which is typical for an indoor environment with no active sources or known pollutant activities occurring. During occupancy, the PNC was significantly higher due to human activity (such as walking on the carpeted floor) and the influence of outdoor sources. The PNC was lowest when the windows were closed, and the office is unoccupied. In this case study, the PNC was also considerably lower when the windows were closed, regardless of human activity, suggesting that a large number of UFPs measured indoors originated from the external environment.

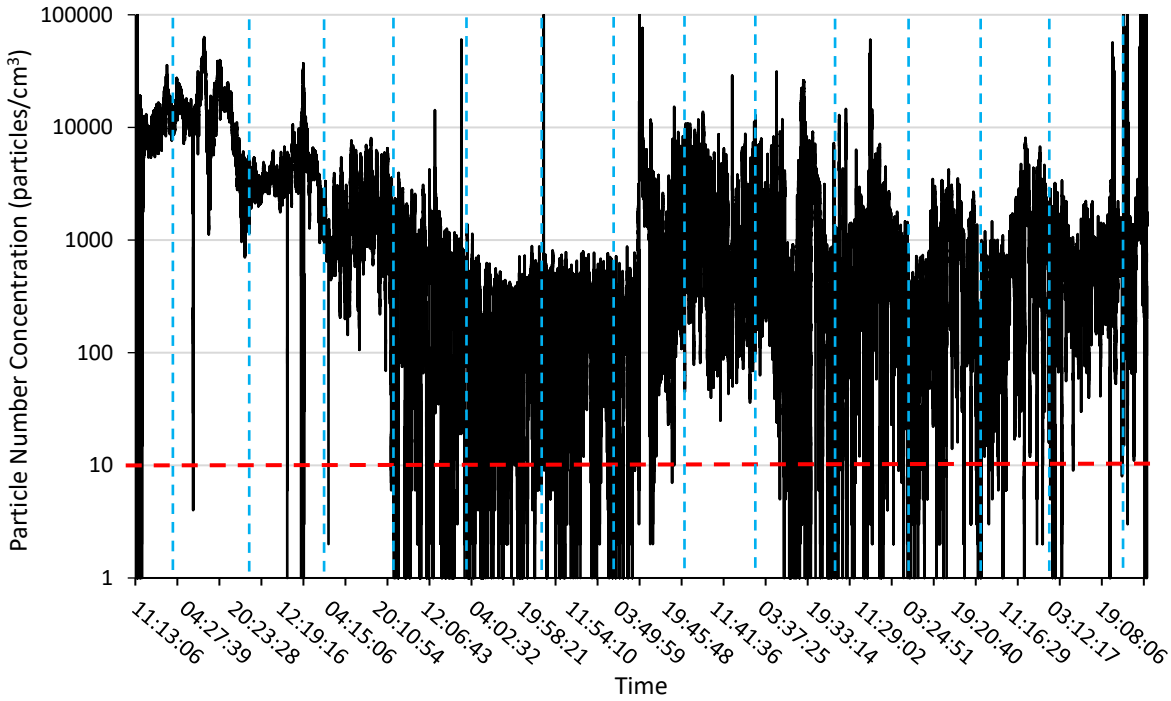


Figure 9.6: Temporal Particle Number Concentrations (particles/cm^3) over the measurement period from 11:13 on 17th July to 11:00 on 31st July. Data is divided up into days of monitoring over the two-week period. PNC scale is logged. Values below dotted line are background concentrations.

Electrochemical Sensors

Although uncalibrated at this point and for this particular study, we can examine the response of the electrochemical sensors to inorganic gaseous pollutants. We particularly wanted to focus on the temporal response rather than the precise readings due to the uncalibrated nature of the sensors. First, it is noticeable that the temperature of the electrochemical sensor fluctuates between 20 and 30 ˚C which may affect the outputs (Figure 9.7). This variation is expected while the sensor is "warming up". However, in this case study the sensor temperature fluctuated throughout the period of measurement.

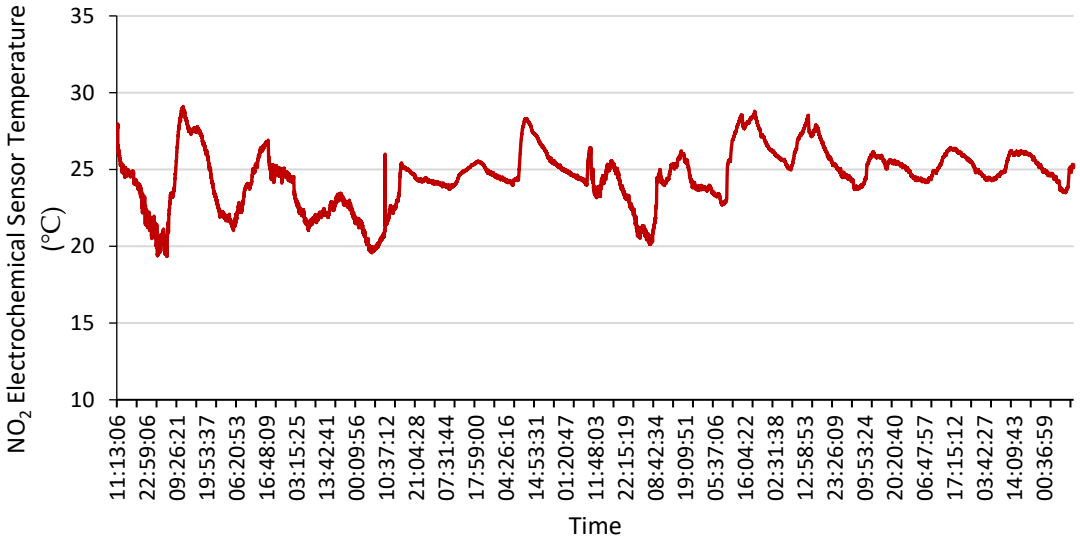


Figure 9.7: Temporal temperature fluctuations (°C) of the NO_2 electrochemical sensor over the full two-week measurement period.

Although uncalibrated, Figure 9.8 illustrates temporal trends in CO, VOC and NO_2 concentrations measured by the electrochemical sensors. In general, temporal fluctuations are small. The large peak in VOC measured during the morning of the 25th July is likely in response to the unit being filled with IPA solution. No significant source of pollution was introduced into the room throughout the measurement period and the three pollutants follow similar trends over time. We do, however, observe some drift in the measurement response which highlights the need for regular calibration if the

sensors were to be used for prolonged periods to enable reporting of reliable results. Some of this drift can also be attributed to sensor warm-up, which has been observed for 48 hours in previous times (such as Lewis et al., 2016).

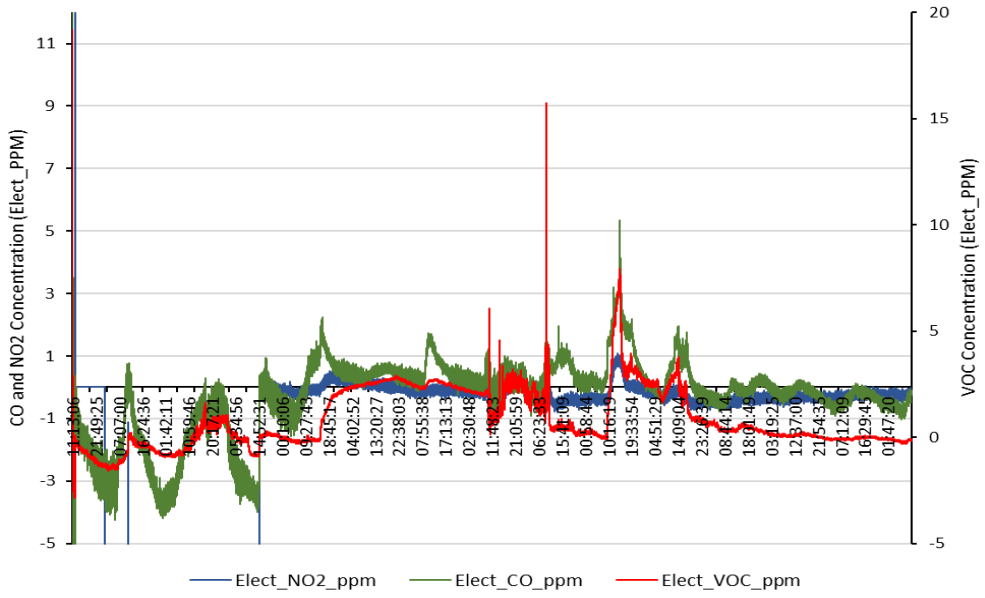


Figure 9.8: Temporal concentrations of Carbon Monoxide (CO), Nitrogen Dioxide (NO_2) and Volatile Organic Compounds (VOC) as TVOC (Total Volatile Organic Compounds) measured by the electrochemical sensors housed in the V1000 units over the entire measurement period.

Case Study 3: VOC Response Case Study (November 2017)

On the 10th November, a case study was undertaken in order to investigate the response of the electrochemical and metal oxide VOC sensors housed in the V1000 units following activation of VOCs in common household products (Figure 9.9). The aim of this case study was to examine the detection capabilities of the sensors, which would inform our understanding of a) the value of these sensors and b) the value of these sensors when linked with active sampling of air and VOC speciation through TD GC-MS. Both sensors have capabilities to quantify TVOC (total volatile organic compounds) so we anticipated seeing a heightened response in measurand (VOC) concentration after activation of a VOC source despite the sensors being uncalibrated at the time of measurement (so we are not confident in the absolute values reported).

This case study was undertaken in an office in LEC 3 that is not normally occupied, and thus is a relatively "clean" environment, free of known strong contaminant sources, to enable us to focus on and characterise the sensor response to the VOCs in common household products.

Products Used;

(1) **Air Freshener** – Air Wick 6 in 1 Air Freshener: Contains Benzisothiazolinone
(2) **Deodorant** – Sure Women's Bright Bouquet Deodorant: Butane, Isobutane, Propane, Aluminium chlorohydrate, Cyclopentasiloxane, PPG-14, Butyl Ether, Parfum, Disteardimonium Hectorite, Propylene Carbonate, Caprylic/Capric Triglyceride, Gelatin Crosspolymer, Cellulose Gum, Sodium Benzoate, Aqua, Hydrated silica, Sodium Starch Octenylsuccinate, Maltodextrin, Hydrolysed Corn Starch, Silica, BHT, Alpha-Isomethyl Ionone, Benzene Alcohol, Benzyl Salicylate, Citronellol, Coumarin, Geraniol, Hexyl Cinnamal, Limonene, Linalool.
(3) **Polish** – Mr Sheen Surface Polish: 5-15% Aliphatic Hydrocarbons, <5% Non-Ionic Surfactants, Perfumes, Geraniol, Limonene, Hexyl Cinnamaldehyde, Preservative, Chloromethylisothiazolinone, Methylisothiazolinone.

Figure 9.9: Products that contain known sources of VOCs (products and specific compounds contained within them listed) that will be activated in an attempt to illicit response by sensors within the V1000 units to evaluate their performance.

The above products are known to contain VOCs and therefore likely to or expected to illicit a response by the (T)VOC sensors in the V1000 units. The chemicals listed on the products have been reproduced here. It is clear to see there are VOCs, sometimes a variety of species, in each product.

VOC Response

Visually, we observe that the VOC electrochemical sensors show the smallest response following the activation of VOC sources at different release distances (Figure 9.10). It is unknown at this stage why this is the case. There is some evidence of response, but this is not clear through interpretation of the data.

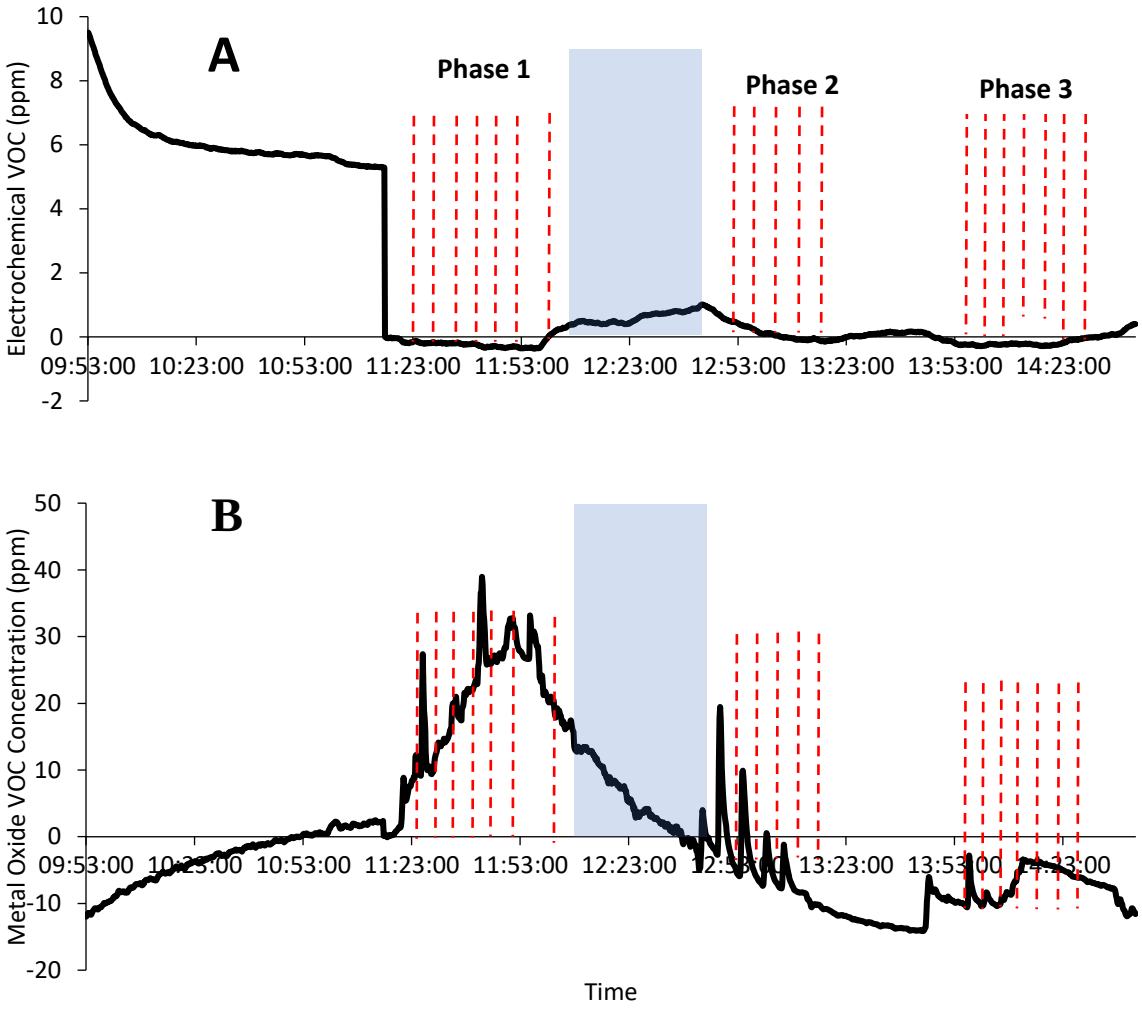


Figure 9.10: VOC response detected by electrochemical VOC sensors (A) and metal oxide (MOS) VOC sensors (B). There are three phases of compound release. In each phase sources were released at increasing distance from the monitoring unit to assess the influence of mixing times. Each dashed line within each phase represents release of products known to contain VOCs and known to illicit a VOC response.

Visually the VOC metal oxide sensor shows a clear response to activation of the VOC sources in all phases (Figure 9.10). There is a heightened peak in VOC concentration following source activation. Sources activated with greater distance from the monitoring unit took longer to reach peak concentrations due to the increased time for

mixing of the air in the room. Sources activated the furthest away from the units incurred the smallest increases in concentration due to dispersion of pollutants.

There is also a clear correspondence by the CO metal oxide sensor. Since this case study occurred in an environment with no known CO sources it is likely this is due to CO sensor cross interferences with VOCs. There is a very high correlation of 0.79 between MOX CO and MOX VOC sensor measurements which is even higher (0.94) if the first hour of "warm-up" measurements are removed. Cross interference on sensor response has similarly been highlighted in the literature (Lewis et al., 2016).

It is difficult to look at comparative statistics in this case study since there was no return to background concentration following the activation of each source. However, statistics can be used to determine whether there was a statistically significant difference in the means of the sensor response before and after activation the first VOC source. This would confirm whether the sensors had detected or responded to activation of the source. A paired t-test was run since the samples are dependent or essentially connected. For the electrochemical sensor the t-stat (calculated t-value) is less (1.68) than the t critical two-tailed (2.05) statistic so we can retain the null hypothesis that states there is no difference between the means of the samples. This highlights that there is no clear response by the electrochemical sensor after initial activation of the source. However, for metal oxide sensors, the t-stat (calculated t-value) is more (16.81) than the t critical two-tailed (2.05) statistic so we can reject the null hypothesis and confirm that there is a statistically significant difference between the means of the samples. This highlights that there is clear response by the MOx sensor following activation of the source. Attributed to sensor drift overtime, and since there is no return to background levels, we cannot perform statistical testing at any other time.

Temporal Trends in CO_2

Temporal trends in CO_2, although not the focus of this case study, were also examined. We can see the clear relationship between CO_2 and occupancy (Figure 9.11). The CO_2 concentration reaches ~ 900 pm during occupancy by one person. Background CO_2 concentration is typically ~ 500 ppm. During periods of non-occupancy CO_2 declines to background levels, with decay rates enhanced when windows are opened to promote air exchange.

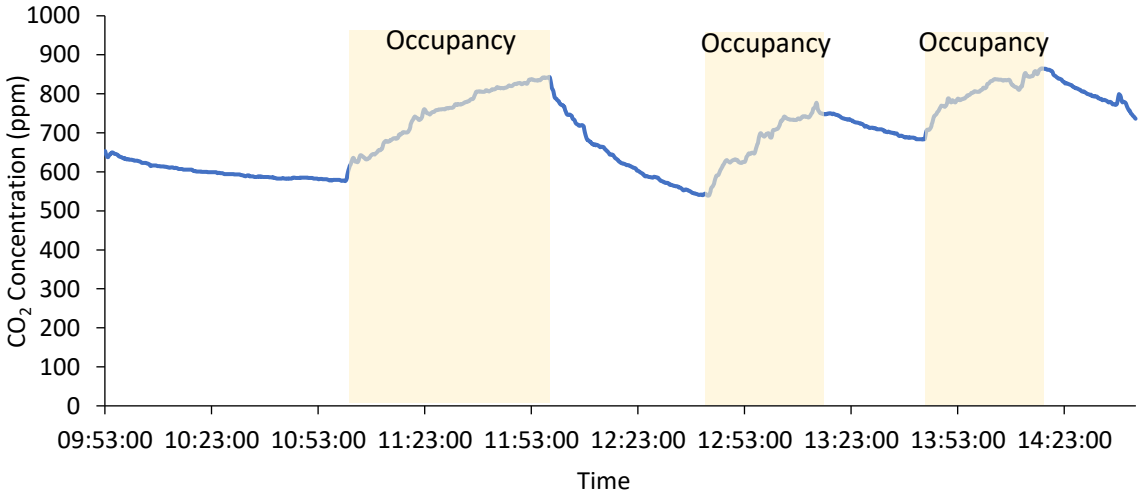


Figure 9.11: Temporal CO_2 concentration fluctuations during the VOC case study and thus activation of VOC sources, with periods of occupancy highlighted (when sources where being activated), that tend to correspond with heighted CO_2 responses.

Air exchange rate was calculated between 12:00 pm and 12:30 pm during a period of non-occupancy. During this time, the windows were open, resulting in approximately 1.6 air changes per hour. This is a relatively high air exchange rate in comparison with those previously calculated from a similar sized office. This is attributed to the fact that both windows were wide open (wider window size opening (larger area for exchange)) and there was a rapid dispersion and decay of CO_2.

Case Study 4: House 5 (February 2018)

This case study was designed to investigate the PNC response to typical household activities as a precursor to generating data from household activities for the thesis (Chapter 4 and 5). While initially it was hoped this case study would continue over multiple days, there was a problem with particle concentration reporting and therefore only two days of reliable data were captured (Figures 9.12 and 9.13). On the first day we can see clear responses to cooking activities in both the PNC response and CO_2 response. We see large peaks in PNC response but there are much more interesting kinetics than these large-scale peaks and troughs, namely, the dynamics of particle response and varying rates of production and removal. We interpret the variability as a function of source characteristics and fluctuations in source strength as well as particle dynamics, most importantly dispersion, but also deposition and coagulation. On the second day we see one short but very clear response to cooking followed by a prolonged period of decline. Baseline PN is usually between 1–10,000 particles/cm^3 on both days.

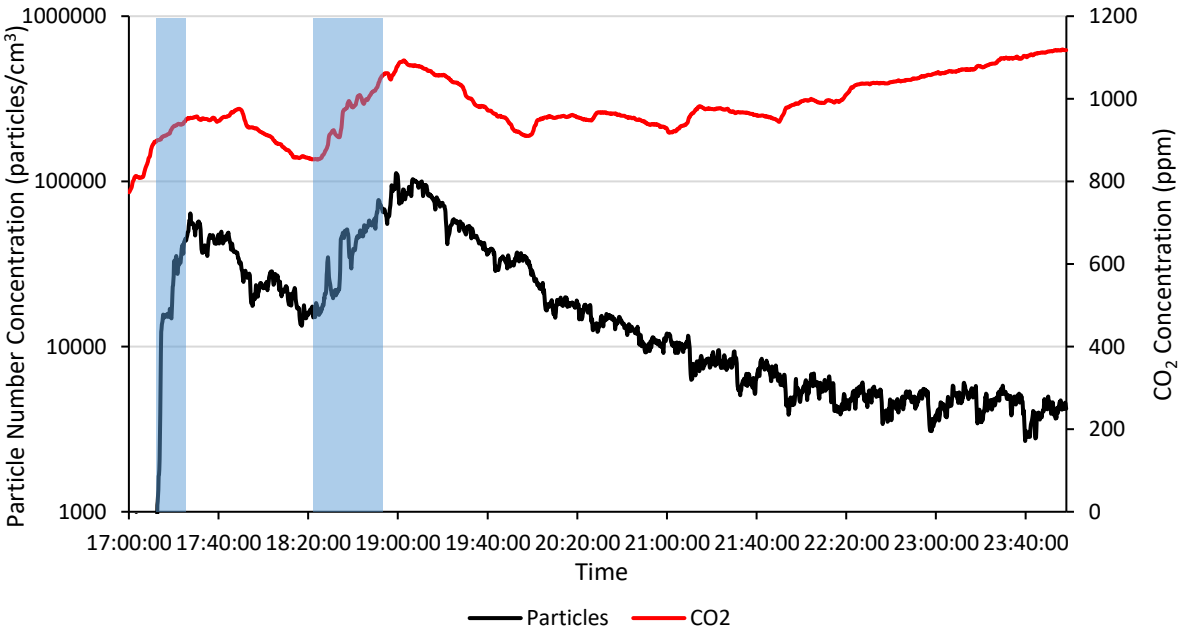


Figure 9.12: Temporal particle number concentration (PNC) response and corresponding CO_2 response to household cooking activities (the undertaking of which is highlighted by shading) on Day 1 of this case study (between 17:00 and 24:00).

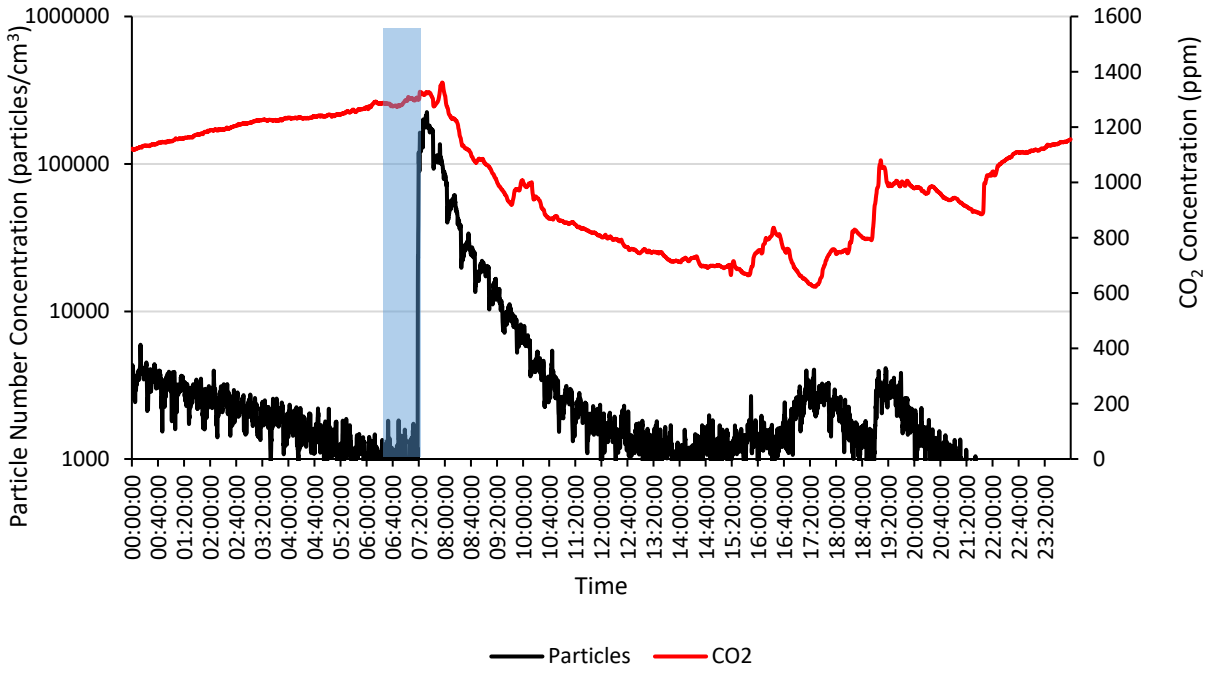


Figure 9.13: Temporal particle number concentration (PNC) response and corresponding CO_2 response to household cooking activities (the undertaking of which is highlighted by shading) on Day 2 of this case study (between 00:00 and 24:00). Sharp rise and slower decline in PNC clearly identified.

In this case study, which used the V1000, we saw the regular filling of the CPC with IPA solution as important. Unless there is an external tank attached to the CPC with additional fluid, we can only expect the units to continue running and reporting reliable PNC data for between 2 and 3 days. NAQTS incorporated these findings into the development of its V2000 device which includes a larger internal tank and other software features that minimise working fluid consumption.

Case Study 5: House 1 – (May 2018)

This case study was designed to investigate PNC response to typical household activities conducted in series (succession) without interspersed periods of ventilation. On reflection this took a similar approach to that taken by the HOMEChem team during their study at the University of Texas (Farmer et al., 2019). Two V1000 monitors were

deployed to capture spatial variation around the residence in question (later termed House 1 in Chapter 4). The monitoring was undertaken over a period of 7 hours (17:00-24:00). The activities were conducted as follows (vacuuming 17:44–17:50, toasting 17:48, oven cooking 18:30 (with the oven door opened at 18:43, 19:04) and egg frying commencing at 19:17). These events cumulatively enhance PNC making it difficult to determine how much each event contributes to PNC given their close occurrence. However, we do see a discrete peak for oven cooking activity that occurred in isolation between 18:30 and 19:04. We observe that there is a steady decline in PNC at the end of each phase of activity (firstly following vacuuming and toasting, and then following oven cooking and egg frying). There is close correspondence in the PNC readings reported by both V1000 monitors, even though one monitor is located downstairs and the other upstairs (Figure 9.14, 9.15). It is also interesting, however, to note that the upstairs V1000 monitor reports a much stronger PNC response despite being further away from most of the sources activated. This we attribute to the influence of housing structure, which we also make reference to in one of the papers after further experimental investigation (Chapter 4). We similarly observe enhanced CO_2 concentrations during periods of cooking activity and occupancy and following declines in response to non-occupancy in the evening (Figure 9.16).

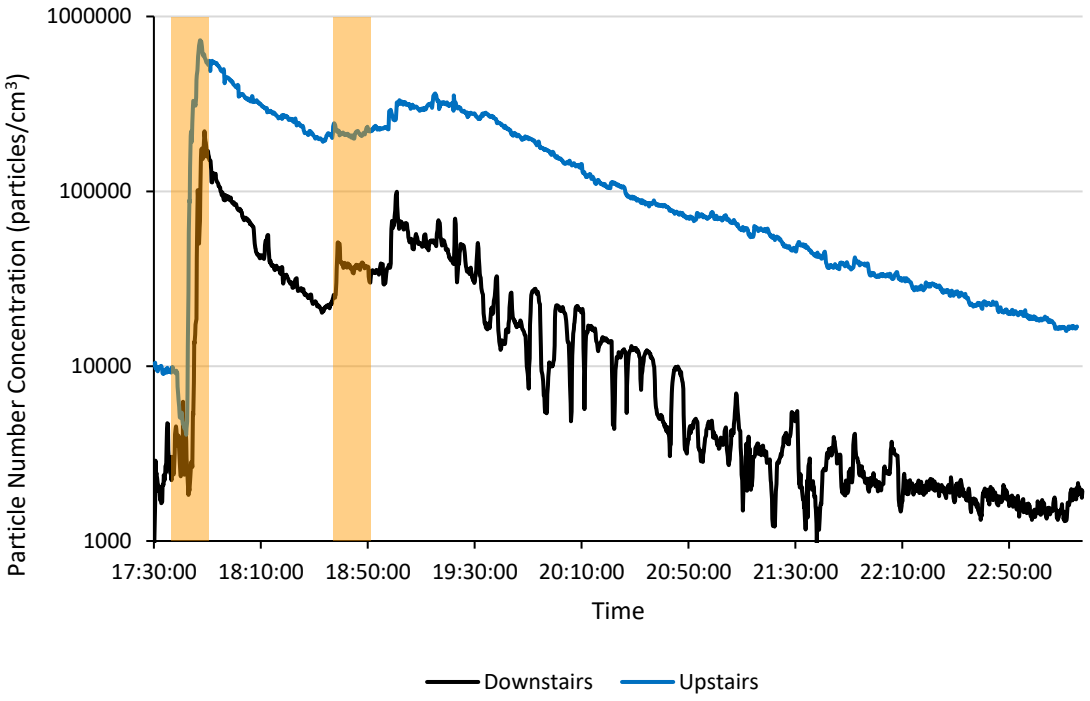


Figure 9.14: Temporal particle number concentration (PNC) response during the undertaking of multiple household activities (vacuuming 17:44–17:50, toasting 17:48, oven cooking 18:30 (with the oven door opened at 18:43, 19:04) and egg frying at 19:17) in a house (later referred to as House 1 in the main thesis) conducted in succession (the occurrence of which is highlighted).

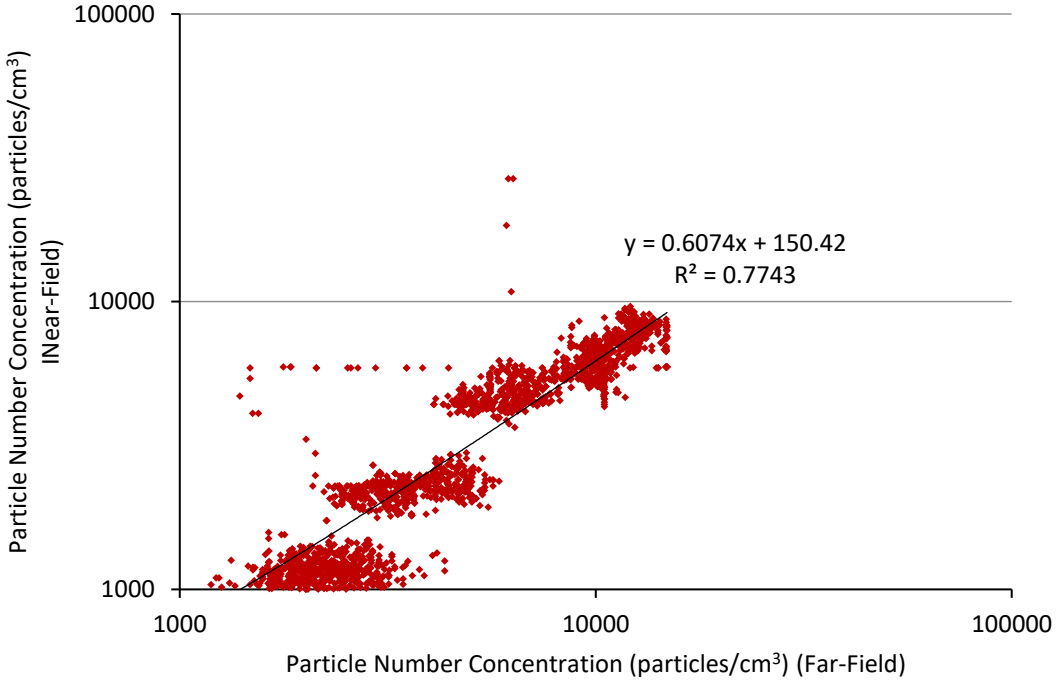


Figure 9.15: Correlation in particle number concentration (PNC) measurements reported by the

two V1000 monitoring units at near field (downstairs) and far field (upstairs) locations in the house investigated.

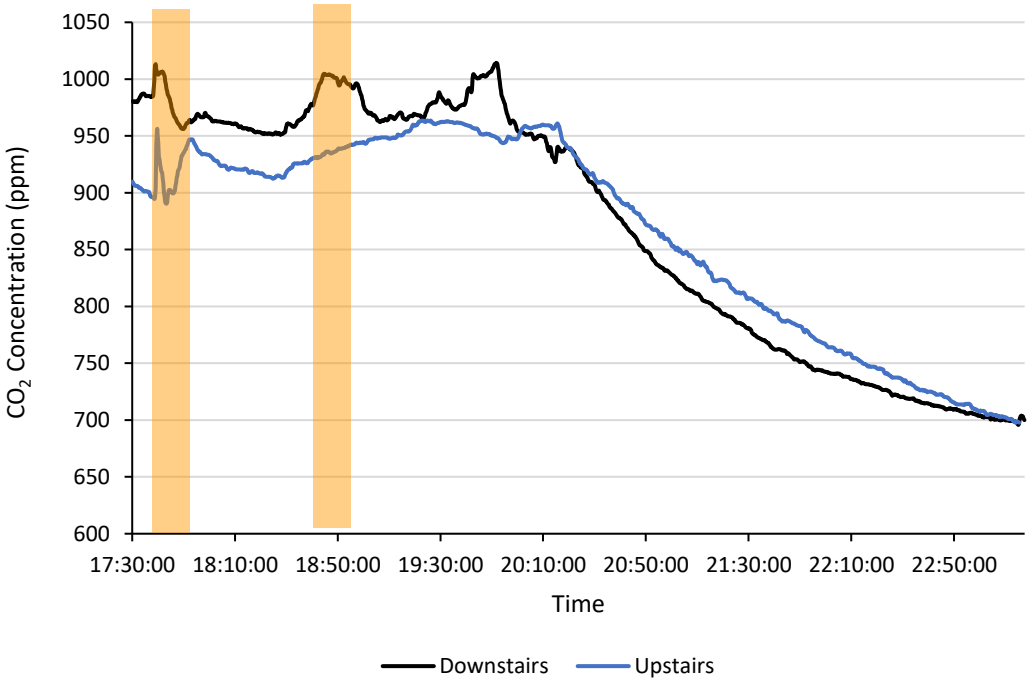


Figure 9.16: Temporal CO_2 concentrations taken simultaneously with the particle measurements that are displayed in Figure 9.15 (between 17:30 and 23:15). The undertaking of household activities previously mentioned are highlighted.

Case Study 6: House 2 - (May 2018)

This case study was designed to test the practicalities of using the NAQTS V1000/2000 monitors in a real-world home situation, and to examine the response of the equipment to ultrafine particles (UFP) generated by episodic toasting events which are interspersed with periods of ventilation and "flushing" between events. This case study was undertaken at a house that was later used in the thesis for further experimental work. One monitor was placed in the kitchen whilst the other was placed upstairs. The scenarios were conducted as follows: A) Toasting (No Ventilation), B) Toasting (Ventilation with Extractor Fan), C) Toasting (Ventilation with Window Opening) and D) Gas Hob (No Ventilation). Differences in source strengths are attributed to differences in toasting activity and variable air exchange provided by ventilation and

the fact that air is not uniformly mixed. Equipment error may play a minor role. We see ventilation significantly enhanced PNC decay rate (Figure 9.17). Mechanical ventilation would not typically be used in conjunction with toasting. However, we wanted to look at the effectiveness of using this strategy to reduce exposure to PNC generated by toasting, despite the distance between the toaster and extractor fan, because we know with increasing airtightness of buildings, mechanical ventilation will become a more common form of ventilation in the future. We see the fastest PNC decay rate when opening windows for enhanced ventilation as expected. In addition to this window opening immediately after toasting appeared to inhibit high peak concentrations being reached.

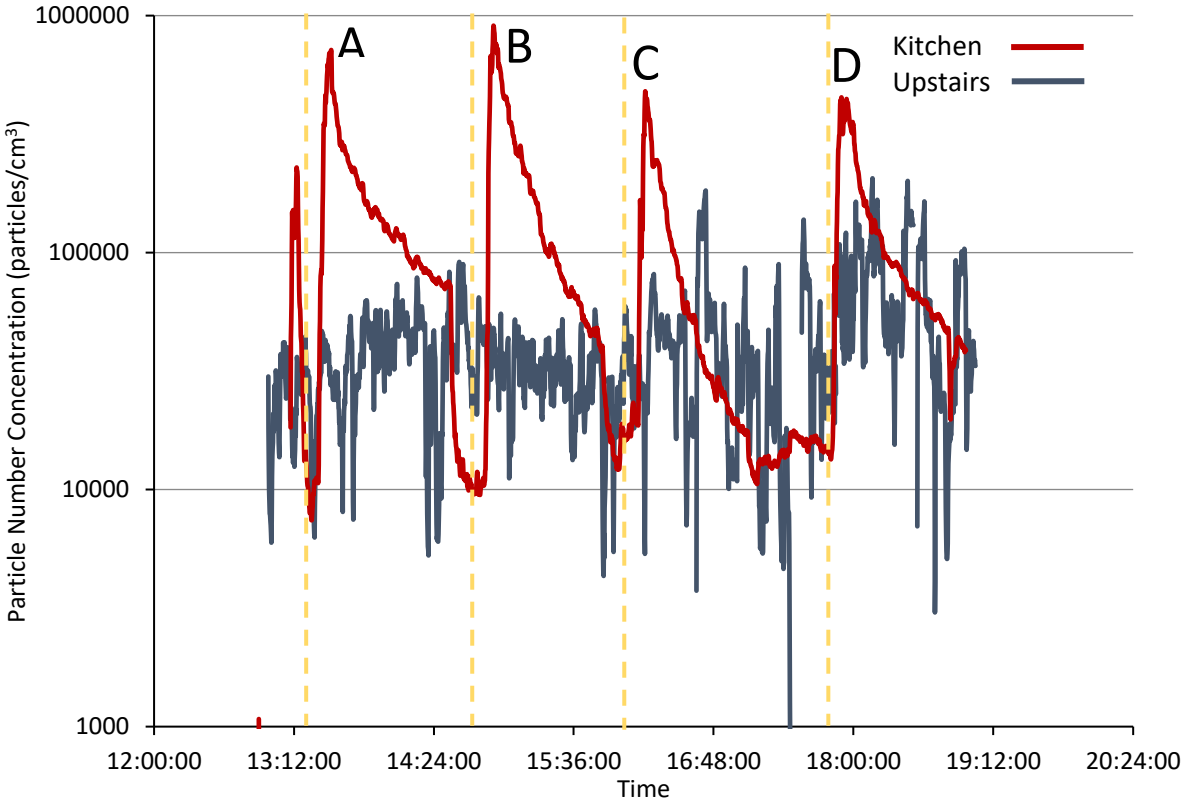


Figure 9.17: Temporal particle response (PNC) from each discrete cooking activity and following ventilation; A) Toasting (No Ventilation), B) Toasting (Ventilation with Extractor Fan), C) Toasting (Ventilation with Window Opening) and D) Gas Hob (No Ventilation) within

the house monitored PNC is measured in the near field (kitchen) and also far field (upstairs) locations to capture spatial variability around the house. Activation of toasting or gas cooking is indicated by dashed lines.

Considering all the toasting activities, we see the smallest range in PNC when ventilating with window opening (Figure 9.18). It is postulated that window opening, through promoting dispersion of toasting generated particulates, suppresses high PNC being reached. The largest range in PNC occurs when the extractor fan is used. This could be attributed to the recycling of particles and pollutants if the extract fan does not vent outside. We observe enhancements in CO_2 in response to cooking activities, with the most significant increase in response to gas cooking at 18:00, as expected (Figure 9.19).

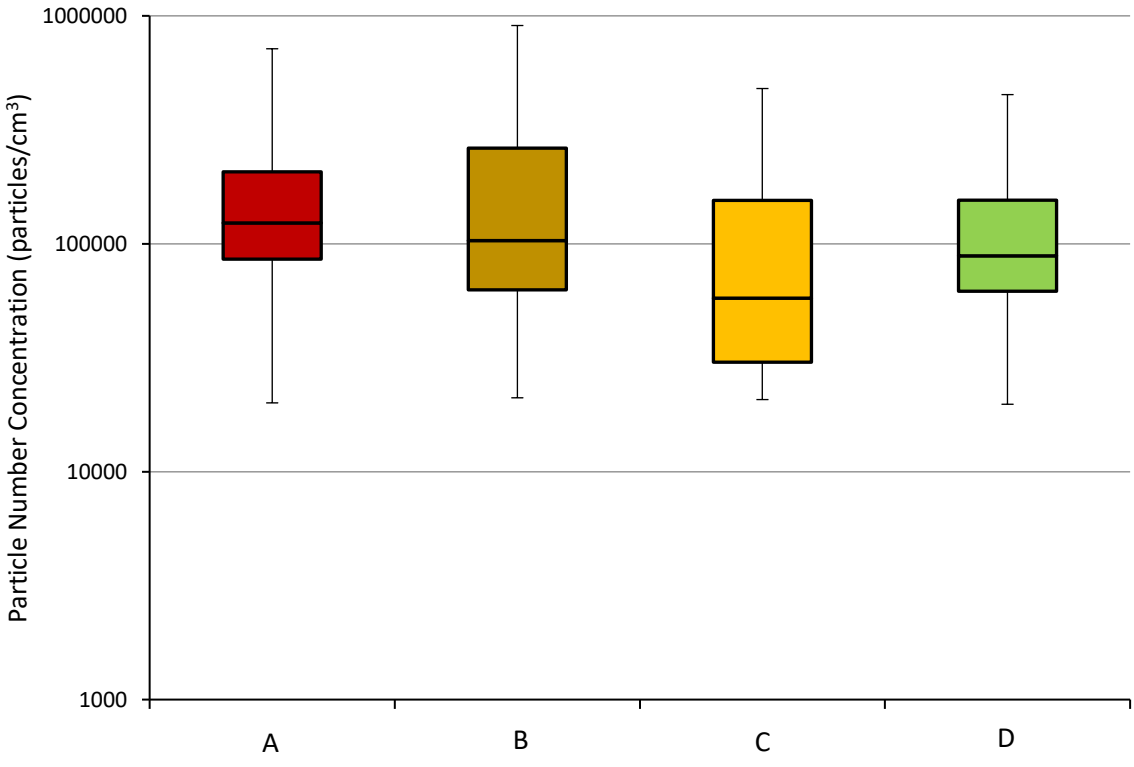


Figure 9.18: Boxplots for each cooking activity illustrating the distribution of particle concentration data. Upper and lower bounds of box represent 25% and 75% of particle data, and the line on each box represents the median value for each event. Upper and lower particle limits reached are also shown for each event.

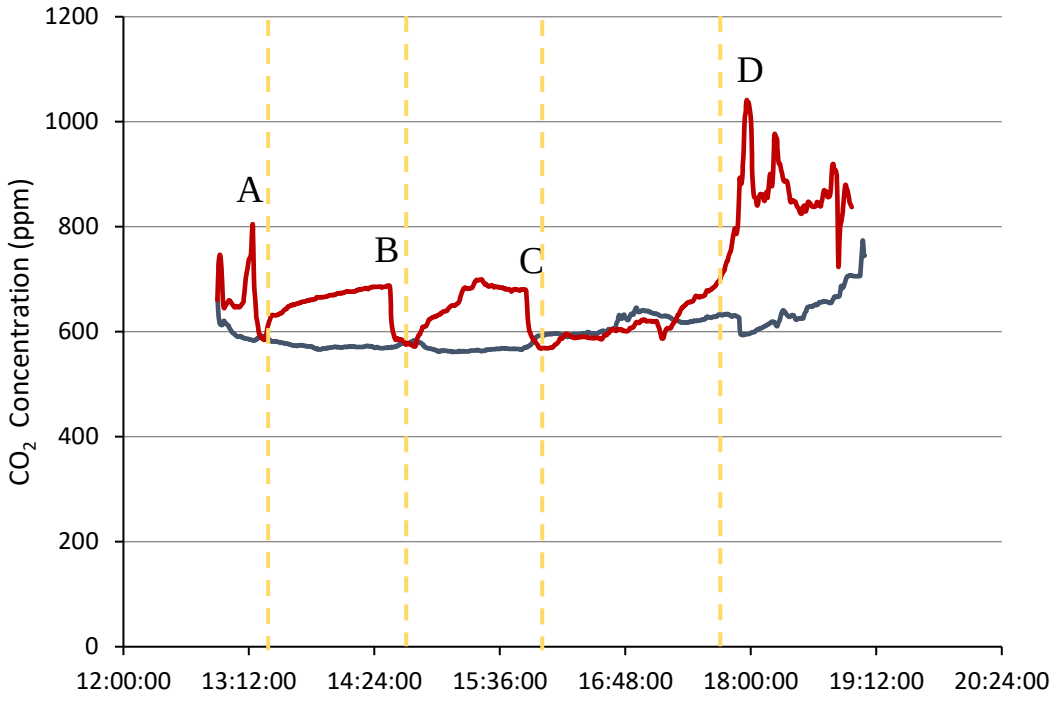


Figure 9.19: Corresponding temporal CO_2 trends from each discrete cooking activity and following ventilation; A) Toasting (No Ventilation), B) Toasting (Ventilation with Extractor Fan), C) Toasting (Ventilation with Window Opening) and D) Gas Hob (No Ventilation). Activation of toasting or gas cooking is indicated by dashed lines.

Case Study 7 – House 2 – (December 2018)

This case study was designed to investigate the application of V1000/2000 monitoring units in the real world with other members of the supervisory team (to also investigate ease of use) using House 2 as a base with a variety of polluting sources. During this case study, combustion-related sources were activated, and the PNC response was examined. Cooking activities were captured at around 18:00 (with the monitor being in the living room). A log burner was then lit in the living room, with the PNC peaks indicating where additional fuel was added to the burner prior to the fire being allowed to burn out (20:00-23:30) (Figure 9.20). Fluctuations in PNC response from the log burner are likely to relate to heterogeneity in burning as we would similarly experience from the burning of a candle in response to heterogeneity in the wick, candle wax or in

the case of scented candles, the added fragrance (Wallace et al., 2019). As the log fire dwindles we see a steady decline to background concentrations. It takes over 4 hours to decay to background PNC. We observe corresponding increases in CO_2 concentration, particularly during the cooking events (Figure 9.21) which remain relatively consistent until the fire burnt out.

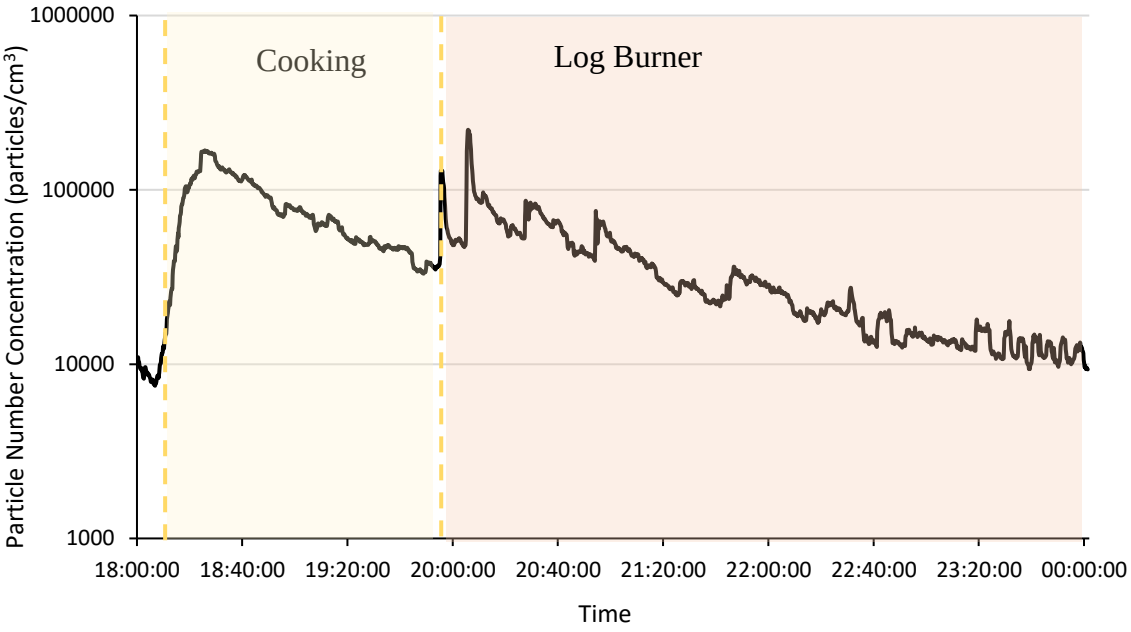


Figure 9.20: Temporal particle number concentration response to combustion related activities; cooking activities in the kitchen (18:15 onwards) and log-burning (19:55 onwards) with monitoring in the living room. Dashed lines indicate start of each activity.

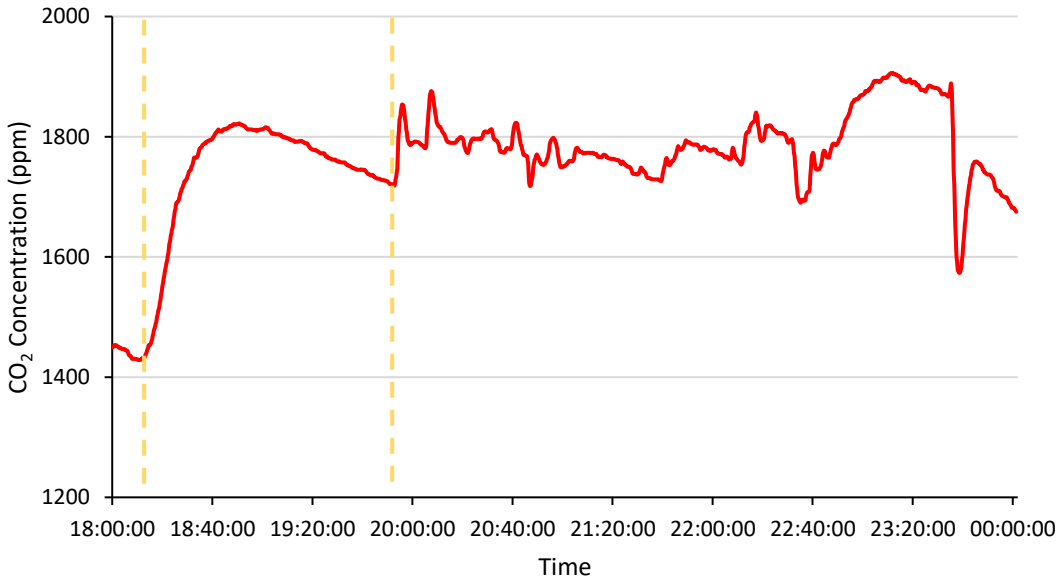


Figure 9.21: Temporal CO_2 measurements corresponding particle number concentration data (Figure 9.20) throughout combustion related activities – cooking activity (18:00 onwards) and operation of a log burner (19:55 onwards). Dashed lines indicate initiation of such activities.

We recognised that the noise of the CPC on the V1000 unit, ~60dB was problematic, being disruptive in the home environment. We also experienced some connectivity issues between the V1000 monitors and computers used to control the data logging process. We therefore decided to conduct subsequent studies during periods of non-occupancy with the sole investigators being the supervisory team and close contacts.

Case Study 8 – House 4- Layered Data Experiments (December 2018)

This case study was undertaken at House 4 and involved testing a protocol that although not subsequently adopted, did inform the ways in which data were eventually collected. It took a layered approach, based on that approach taken during by the HOMEChem project (Farmer et al. 2019), to assess PNC response to various cooking activities throughout the day under periods of no ventilation (Figure 9.22) interspersed with periods of ventilation (Figure 9.23). Again, two V1000 monitors were deployed, one at a near-field location close to the source (kitchen) and another at a far-field location (upstairs). We observe significant enhancements of PNC in response to each individual event, and persistence over long periods under no ventilation resulting in significant concentrations but consistently lower concentrations for the far-field monitor (located further from the source). We cannot discern the specific influence of individual sources completely due to the interactions and overlapping nature of the experiments, but this is nonetheless important to understanding exposure in the real-world where sources will interact and overlap. We observe much more rapid declines in PNC response during interspersed periods of ventilation (window opening) and lower exposures. During

ventilation we often saw PNC return to background levels before activation of another source. However, this was not the case under conditions of no ventilation.

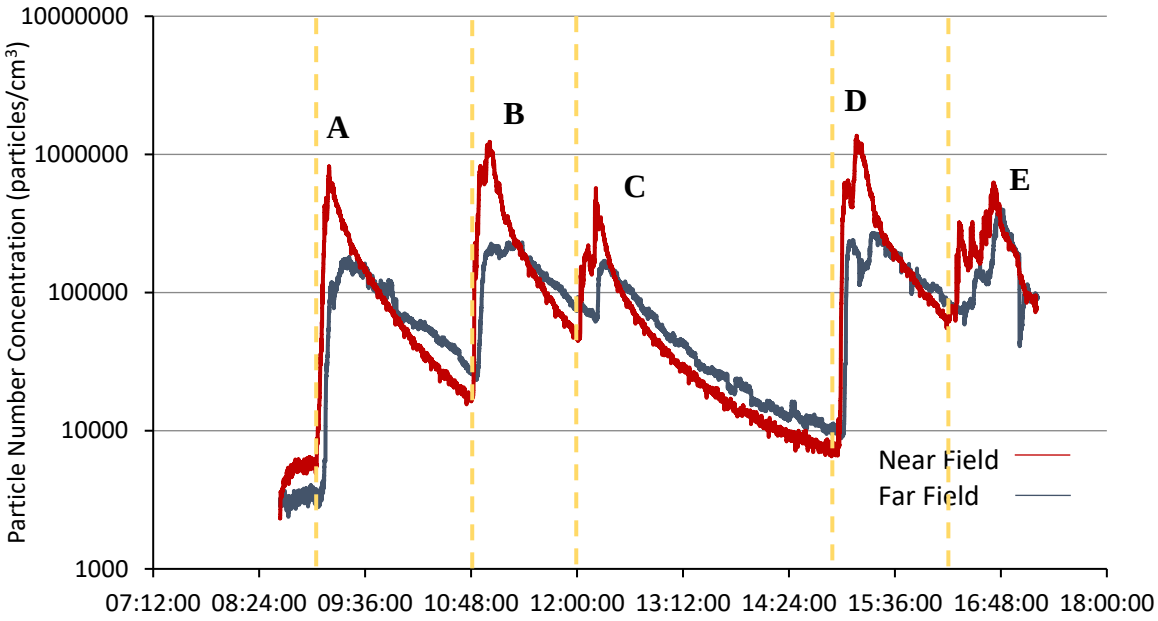


Figure 9.22: Temporal particle number concentration (PNC) response from typical household cooking activities under no ventilation. (A) Toasting; (B) Frying; (C) Microwave and (D and E) More complex dinnertime cooking involving frying and oven cooking. Activities are conducted consequentially without necessitating a return to background levels. Dashed lines indicate source activation i.e., the start of cooking activity.

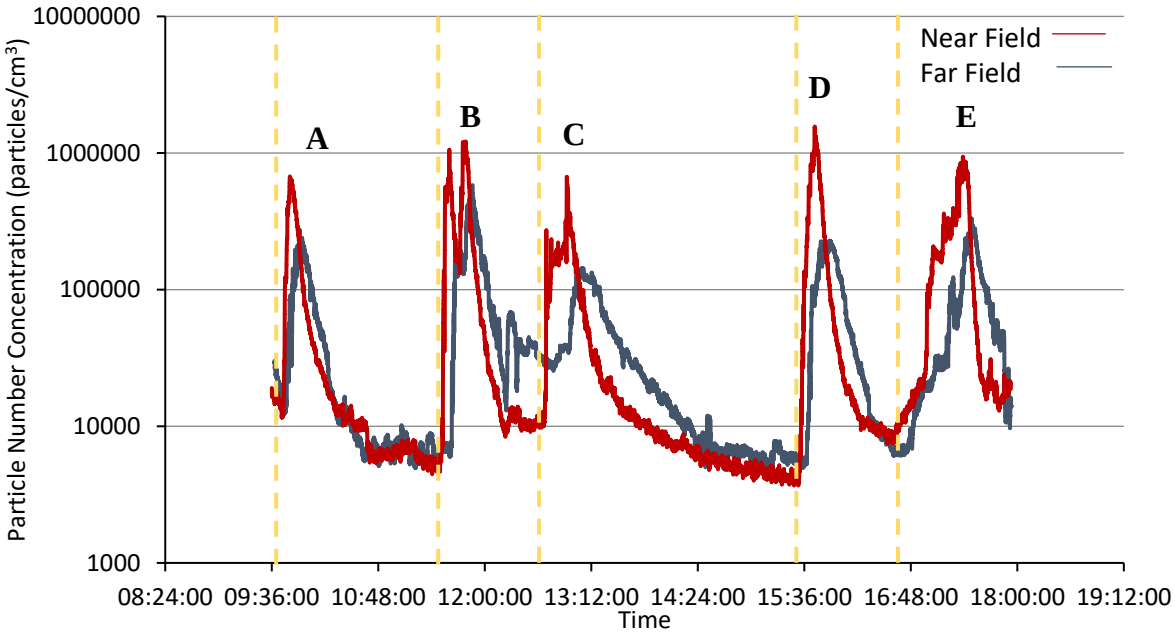


Figure 9.23: Temporal particle number concentration (PNC) response from typical household activities under a naturally ventilated regime (each activity is interspersed with periods of ventilation i.e., window opening). The same activities are repeated as previously; (A) Toasting; (B) Frying; (C) Microwave and (D and E) More complex dinnertime cooking involving frying and oven cooking. Activities are conducted consequentially without necessitating a return to background levels. Dashed lines indicate source activation i.e., the start of cooking activity.

Summary

This chapter has presented several case studies showing the application of NAQTS V1000/V2000 monitoring units in real-world settings. These V1000/V2000 air quality monitoring units have been tested here for their effectiveness and accuracy largely in detecting and representing typical household cooking activities. These case studies informed the way we conducted our research in the main body of the thesis. They inform us about the use of the units in Chapters 4 and 5, and to a lesser extent, Chapter 6. These units have multiple pollutant monitoring capabilities, but we largely focus on the PNC response given by these units which is determined by the CPC housed inside the units. When carrying out research throughout this thesis we use these units to investigate the particulate response to typical household cooking activities within real-life and controlled kitchen environments. We focus on PNC response due to the confirmed accuracy and confidence in the reported results. We do not rely or use the electrochemical or metal oxide sensor data. As the literature review discusses, despite the opportunities raised by the use of "low-cost" sensor technologies, there are many problems associated with them, even though there have been some attempts to compare against reference instrumentation. We observed significant drift in sensor response in the case studies, which would mean that the sensors would require frequent multi-parameter calibration for us to be confident in reporting absolute values. The complexities of this calibration are at odds with the aims of the thesis which aims to

promote widespread public understanding of IAQ and gain high time and spatially resolved data. However, this sensor technology does hold some promise for reporting IAQ, and we should not discount their potential in the future for being able to reliably report pollutant concentrations at a great spatial and temporal resolution.

We therefore conclude;

- PNC: CPC was calibrated by Ricardo AEA (according to ISO 27891) and therefore provides high quality, high temporal resolution PNC data. V1000/V2000 Units were collocated to normalise data between them for increased accuracy. We are confident in the PNC data provided for studies on cooking and ventilation in domestic and specialised settings. Outliers can be removed prior to analysis by flag detection.
- CO_2 NDIR sensor – provides a useful measure and indicator of occupancy and used to calculate air exchange rates.
- VOC – not used in this thesis (as uncalibrated, and not seen to be confident in results) – however sensor technology shows promise for VOC detection and understanding temporal signatures (which we allude to in Chapter 6) especially when integrated with VOC speciation (via thermal desorption tubes, which can be attached to the units for time integrated sampling).

Based on these case studies we decided to further evaluate discrete cooking activities for 3 easily replicable sources (toast, eggs, bacon) under conditions of no ventilation, natural ventilation, and mechanical ventilation in sequential experiments (to better enable source characterisation). Flushing between events enabled us to easily separate events (and understanding individual event characteristics and exposures) whereas layered experiments (closer to mimicking real-life) were harder to interpret due to added

complexities. The case studies reported in this technical evaluation also helped us develop a protocol for sampling around a residence. We deployed multiple V1000/V2000 units in near and far field locations in a single room (kitchen) and around a residence. We are confident in the reporting abilities of the units, made through correlations between them. The protocol established to run these monitors and their locations for sampling has informed the protocol and decisions for subsequent sampling. We found problems associated with the units which included noise and practicalities of the user interface. Therefore, we ultimately made the decision to restrict sampling to be undertaken by members of the supervisory team and not the wider public. The problems with noise have since been reduced in adaptations made to the units.

Appendix A1: Key Metrics Reported from Data in Chapter 3 (Supplementary Information)

In this section we present the metrics devised from the raw temporal particulate data reported from two monitors in one room (Kitchen – Near and Far to Source).

Table S1: Area-under-the-curve (AUC), which is taken as a cumulative total of pollutants emitted and surrogate of source strength, for near-field (kitchen) location, reported to 3 significant figures (x 10^8 cm^3).

	H1	H2	H3	H4	H5	H6	H7	H8
Toast No Ventilation	5.04	8.61	12.2	9.75	1.48	5.91	9.64	8.77
Toast Natural Ventilation	0.63	1.31	1.12	3.36	0.19	1.45	0.21	3.93
Eggs No Ventilation	1.26	17.5	21.4	19.4	5.48	15.8	5.59	14.3
Eggs Mechanical Ventilation	2.17	6.74	12.6	21.2	1.02	8.62	2.17	10.6
Eggs Natural Ventilation	2.19	2.23	8.34	9.58	0.60	3.13	1.68	5.75
Bacon No Ventilation	12.5	6.46	15.7	4.28	2.02	3.72	13.6	24.1
Bacon Mechanical Ventilation	15.3	4.18	7.21	7.65	0.65	2.86	10.6	17.8
Bacon Natural Ventilation	1.07	1.34	3.88	3.78	0.54	1.43	8.54	15.5
Mean	5.03	6.05	10.3	9.86	1.50	5.36	6.50	12.6

Table S2: Area-under-the-curve (AUC), which is taken as a cumulative total of pollutants emitted and surrogate of source strength, for far-field (kitchen) location, reported to 3 significant figures (x 10^8 cm^3).

	H1	H2	H3	H4	H5	H6	H7	H8
Toast No Ventilation	9.20	10.7	9.22	6.82	5.47	6.83	13.2	9.76
Toast Natural Ventilation	0.95	9.31	0.58	1.47	0.87	1.54	3.57	5.88
Eggs No Ventilation	0.64	18.2	10.9	12.9	17.6	15.2	7.79	15.4
Eggs Mechanical Ventilation	1.07	4.84	6.68	14.8	3.50	7.51	2.56	10.8
Eggs Natural Ventilation	1.22	2.04	3.27	4.47	2.47	3.81	2.63	8.62
Bacon No Ventilation	11.6	5.23	9.61	3.95	7.02	2.26	5.44	20.6
Bacon Mechanical Ventilation	15.1	2.15	6.38	7.24	1.50	1.02	3.83	16.8
Bacon Natural Ventilation	9.87	0.76	2.32	2.60	2.03	0.53	1.80	13.4
Mean	6.20	6.65	6.12	6.78	5.06	4.84	5.10	12.65

Table S3: Time to background (TTB) metric, which is a measure of the time from peak particle concentration to background particle concentration for near-field kitchen location (min).

	H1	H2	H3	H4	H5	H6	H7	H8
Toast No Ventilation	133	210	183	120	95	67	160	77
Toast Natural Ventilation	12	41	35	34	17	16	47	29
Eggs No Ventilation	79	218	150	172	218	116	88	82
Eggs Mechanical Ventilation	133	112	78	126	43	59	61	63
Eggs Natural Ventilation	73	31	39	40	18	17	33	23
Bacon No Ventilation	242	108	208	60	101	54	112	78
Bacon Mechanical Ventilation	210	67	105	83	60	51	84	57
Bacon Natural Ventilation	83	42	31	20	8	8	24	29
Mean	121	104	104	82	70	49	76	55

Table S4: Time to background (TTB) metric, which is taken as a measure of the time from peak particle concentration to background particle concentration for far-field kitchen location (min).

	H1	H2	H3	H4	H5	H6	H7	H8
Toast No Ventilation	193	216	166	119	163	70	162	72
Toast Natural Ventilation	22	37	30	31	33	17	43	27
Eggs No Ventilation	47	197	123	167	226	100	74	74
Eggs Mechanical Ventilation	50	94	78	124	67	69	65	56
Eggs Natural Ventilation	49	27	37	39	29	18	33	22
Bacon No Ventilation	252	109	200	60	140	30	108	79
Bacon Mechanical Ventilation	147	83	98	86	50	30	75	60
Bacon Natural Ventilation	83	26	30	16	35	6	15	29
Mean	105	99	95	80	93	43	72	52

Table S5: Time to peak (TTP) metric, which is taken as a measure of the time from the start of activity until peak particle concentrations are reached for near-field kitchen location (min).

	H1	H2	H3	H4	H5	H6	H7	H8
Toast No Ventilation	7	11	12	5	4	5	8	5
Toast Natural Ventilation	7.5	10	5	4	5	4	8	6
Eggs No Ventilation	14	11	8	8	8	7	9	7
Eggs Mechanical Ventilation	9	5	6	5	7	23	12	8
Eggs Natural Ventilation	14	8	6	7	4	6	5	10
Bacon No Ventilation	21	24	16	20	9	18	21	19
Bacon Mechanical Ventilation	10	24	13	10	2	22	20	20
Bacon Natural Ventilation	22	22	21	21	18	20	20	19
Mean	13	14	11	10	7	13	13	12

Table S6: Time to peak (TTP) metric, which is taken as a measure of the time from the start of activity until peak particle concentrations are reached for far-field kitchen location (min).

	H1	H2	H3	H4	H5	H6	H7	H8
Toast No Ventilation	7	18	12	7	10	5	4	3
Toast Natural Ventilation	8	5	4	5	10	5	3	4
Eggs No Ventilation	12	28	10	9	13	8	14	6
Eggs Mechanical Ventilation	11	10	6	8	10	22	8	7
Eggs Natural Ventilation	13	9	9	9	9	6	6	9
Bacon No Ventilation	11	23	20	23	10	19	22	23
Bacon Mechanical Ventilation	13	23	16	11	18	23	24	20
Bacon Natural Ventilation	22	23	22	26	20	23	22	21
Mean	12	17	12	12	13	14	13	12

Table S7: The rate of decay (RTE) metric, which is taken as a measure of the rate of particle decay from peak to background, for near-field kitchen location, reported to 3 s.f. (cm^3/s).

	H1	H2	H3	H4	H5	H6	H7	H8
Toast No Ventilation	8.98	6.41	10.3	19.6	9.07	26.9	9.98	35.1
Toast Ventilation	99.9	37.1	36.2	71.7	35.0	91.6	51.5	104
Eggs No Ventilation	11.4	9.80	16.6	12.5	7.54	17.8	22.9	37.2
Eggs Mechanical Ventilation	9.97	18.0	31.4	15.7	31.6	41.4	19.6	46.5
Eggs Natural Ventilation	17.6	44.7	76.4	73.0	77.6	120	45.8	161
Bacon No Ventilation	8.47	11.3	10.1	23.5	10.6	32.8	16.0	36.7
Bacon Mechanical Ventilation	8.40	26.6	17.5	20.8	30.0	47.6	23.5	45.8
Bacon Natural Ventilation	29.8	28.5	73.3	110	68.9	222	116	104
Mean	24.3	22.8	34.0	43.3	33.7	75.0	38.16	71.4

Table S8: The rate of decay (RTE) metric, which is taken as a measure of the rate of particle decay from peak to background, for far-field kitchen location, reported to 3 s.f. (cm^3/s).

	H1	H2	H3	H4	H5	H6	H7	H8
Toast No Ventilation	8.51	7.08	10.8	18.2	8.35	25.6	9.90	37.2
Toast Ventilation	99.9	39.5	43.1	61.0	34.7	97.8	42.8	124
Eggs No Ventilation	15.6	8.32	13.3	10.5	7.10	15.9	23.7	38.1
Eggs Mechanical Ventilation	21.8	17.1	27.2	13.3	29.5	38.0	22.4	49.1
Eggs Natural Ventilation	21.3	2.98	63.6	63.1	75.6	137	53.1	164
Bacon No Ventilation	8.21	11.2	9.69	16.3	9.78	29.6	11.3	26.2
Bacon Mechanical Ventilation	8.22	22.4	18.8	18.8	25.6	14.9	19.0	49.2
Bacon Natural Ventilation	28.0	31.2	75.0	106	43.7	372	98.9	114
Mean	26.4	17.5	32.7	38.4	29.3	91.4	35.1	75.2

Appendix A2: Temporal particulate metrics as above for near and far-field monitors in two rooms

Table S9: Area-under-the-curve (AUC), which is taken as a cumulative total of pollutants emitted and surrogate of source strength, for near-field (kitchen) location, reported to 3 significant figures (x 10^8 cm^3).

	H1	H2	H3	H4	H5	H6	H7	H8
Toast No Ventilation	3.30	3.58	6.82	7.71	10.2	5.22	6.85	6.88
Toast Natural Ventilation	2.14	1.39	3.18	2.98	4.64	1.94	1.60	2.83
Eggs No Ventilation	7.10	4.97	11.9	10.8	1.66	13.2	6.85	7.56
Eggs Mechanical Ventilation	4.83	6.29	5.37	9.24	0.56	6.27	2.03	5.09
Eggs Natural Ventilation	3.55	2.65	4.10	2.80	0.41	9.38	0.74	4.70
Bacon No Ventilation	22.6	10.6	8.81	5.45	1.69	6.31	14.4	16.1
Bacon Mechanical Ventilation	19.2	7.21	12.6	4.74	0.36	7.19	10.9	16.3
Bacon Natural Ventilation	17.1	4.22	4.87	2.14	0.21	5.27	13.9	13.9
Mean	10.0	5.11	7.21	5.73	2.47	6.85	7.16	9.17

Table S10: Area-under-the-curve (AUC), which is taken as a cumulative total of pollutants emitted and surrogate of source strength, for far-field monitor (upstairs), reported to 3 significant figures (x 10^8 cm^3).

	H1	H2	H3	H4	H5	H6	H7	H8
Toast No Ventilation	8.14	3.42	3.11	4.43	2.22	2.29	4.46	0.71
Toast Natural Ventilation	3.19	0.93	1.17	0.83	0.49	0.71	0.79	2.15
Eggs No Ventilation	4.35	7.41	6.72	7.91	2.26	N/A	3.32	4.24
Eggs Mechanical Ventilation	1.78	1.89	0.89	6.53	0.98	2.65	1.09	1.96
Eggs Natural Ventilation	2.56	3.26	2.83	1.82	1.23	1.30	0.44	4.22
Bacon No Ventilation	7.58	2.84	4.53	3.39	2.43	1.85	5.65	5.11
Bacon Mechanical Ventilation	10.4	2.23	4.49	3.52	0.52	3.12	3.39	3.15
Bacon Natural Ventilation	8.67	1.31	2.26	1.93	1.02	3.53	1.98	7.55
Mean	5.83	2.91	3.25	3.80	1.39	1.93	2.64	3.64

Table S11: Time to background (TTB) metric, which is a measure of the time from peak particle concentration to background particle concentration for near-field location (min).

	H1	H2	H3	H4	H5	H6	H7	H8
Toast No Ventilation	120	142	127	90	150	85	120	65
Toast Ventilation	50	72	39	30	39	26	28	22
Eggs No Ventilation	198	82	127	120	68	93	138	32
Eggs Mechanical Ventilation	146	115	86	93	35	74	63	62
Eggs Natural Ventilation	85	46	48	30	10	16	21	30
Bacon No Ventilation	198	179	120	78	41	60	120	88
Bacon Mechanical Ventilation	184	116	103	60	25	49	102	67

Bacon Natural Ventilation	222	38	26	28	16	27	55	43
Mean	150	98.8	84.5	66.1	48.0	53.8	80.9	51.1

Table S12: Time to background (TTB) metric, which is a measure of the time from peak particle concentration to background particle concentration for far-field (upstairs) location (min).

	H1	H2	H3	H4	H5	H6	H7	H8
Toast No Ventilation	230	154	97	90	137	84	118	55
Toast Ventilation	78	55	38	21	37	24	25	42
Eggs No Ventilation	185	216	133	110	70	N/A	133	81
Eggs Mechanical Ventilation	125	98	60	90	41	84	62	109
Eggs Natural Ventilation	104	95	48	28	16	28	18	80
Bacon No Ventilation	191	135	89	76	56	48	120	93
Bacon Mechanical Ventilation	181	100	87	55	16	46	88	61
Bacon Natural Ventilation	171	27	25	28	12	33	44	70
Mean	158	110	72.1	62.3	48.1	43.4	76.0	73.9

Table S13: Time to peak (TTP) metric, which is taken as a measure of the time from the start of activity until peak particle concentrations are reached for near-field location (min).

	H1	H2	H3	H4	H5	H6	H7	H8
Toast No Ventilation	8	10	4	9	7	8	12	5
Toast Ventilation	3	3	3	4	8	7	7	5
Eggs No Ventilation	11	7	7	7	7	8	14	6
Eggs Mechanical Ventilation	10	7	15	4	11	9	15	6
Eggs Natural Ventilation	11	12	9	5	6	5	12	6
Bacon No Ventilation	15	21	23	21	22	19	21	22
Bacon Mechanical Ventilation	22	17	22	20	18	21	22	22
Bacon Natural Ventilation	16	20	25	18	9	21	25	23
Mean	12.0	12.0	13.5	11.0	11.0	12.3	16.0	11.9

Table S14: Time to peak (TTP) metric, which is taken as a measure of the time from the start of activity until peak particle concentrations are reached for far-field (upstairs) location (min).

	H1	H2	H3	H4	H5	H6	H7	H8
Toast No Ventilation	4	45	24	15	20	12	11	19
Toast Natural Ventilation	5	30	15	8	11	13	7	14
Eggs No Ventilation	11	8	20	15	33	N/A	15	18
Eggs Mechanical Ventilation	19	25	54	10	16	12	12	32
Eggs Natural Ventilation	12	24	13	9	12	11	11	13
Bacon No Ventilation	25	49	49	26	44	30	31	32

Bacon Mechanical Ventilation	25	37	39	23	29	24	32	29
Bacon Natural Ventilation	24	33	34	26	23	21	28	33
Mean	15.6	31.4	31.0	16.5	23.5	15.4	18.4	23.8

Table S15: The rate of decay (RTE) metric, which is taken as a measure of the rate of particle decay from peak to background, for near-field kitchen location, reported to 3 s.f. (cm^3/s).

	H1	H2	H3	H4	H5	H6	H7	H8
Toast No Ventilation	6.66	10.5	10.7	21.0	9.71	24.5	11.4	35.8
Toast Natural Ventilation	27.9	11.8	81.2	62.8	47.0	102	77.1	79.2
Eggs No Ventilation	9.11	17.9	10.3	14.5	18.8	22.3	8.96	105
Eggs Mechanical Ventilation	11.5	17.9	18.09	15.9	25.5	28.4	16.2	35.7
Eggs Natural Ventilation	11.2	41.5	48.1	79.0	116	97.0	76.6	90.1
Bacon No Ventilation	12.5	12.5	11.8	17.9	42.6	27.6	15.6	27.4
Bacon Mechanical Ventilation	12.8	14.5	18.1	25.2	40.1	48.7	16.6	40.1
Bacon Natural Ventilation	10.5	12.5	74.8	65.2	57.7	65.7	26.1	71.9
Mean	12.8	17.4	34.1	37.7	44.7	52.0	31.1	60.7

Table S16: The rate of decay (RTE) metric, which is taken as a measure of the rate of particle decay from peak to background, for far-field location (upstairs), reported to 3 s.f. (cm^3/s).

	H1	H2	H3	H4	H5	H6	H7	H8
Toast No Ventilation	10.1	6.87	7.79	17.0	4.72	9.80	9.97	2.44
Toast Ventilation	23.3	12.6	25.9	50.8	12.5	27.0	63.3	20.3
Eggs No Ventilation	8.15	8.32	12.6	11.7	10.2	N/A	6.83	11.8
Eggs Mechanical Ventilation	7.17	6.45	4.54	14.2	13.1	12.4	12.5	13.3
Eggs Natural Ventilation	11.2	16.1	32.4	65.3	100	57.2	60.8	14.7
Bacon No Ventilation	9.68	6.16	9.17	16.4	24.1	25.6	11.5	14.6
Bacon Mechanical Ventilation	10.3	6.82	13.3	23.9	16.1	29.4	14.7	16.2
Bacon Natural Ventilation	10.6	6.16	44.4	54.5	109	35.2	24.0	23.3
Mean	11.3	8.69	18.8	31.7	36.2	24.6	25.5	14.6

Table S17: The peak concentration (PKC) or maximum concentration metric, which is taken as a measure of the significance of pollutant source or source strength, near to source in kitchen to 3 s.f. (x 10^5 particles/cm^3).

	H1	H2	H3	H4	H5	H6	H7	H8
Toast No Ventilation	1.84	3.42	9.25	10.2	3.93	7.48	3.20	9.77
Toast Natural Ventilation	2.87	7.07	7.21	6.69	9.65	7.68	2.82	10.5
Eggs No Ventilation	4.06	3.27	14.4	11.3	1.49	15.5	3.12	16.9
Eggs Mechanical Ventilation	5.29	5.97	3.18	17.7	0.48	9.18	1.36	9.24
Eggs Natural Ventilation	2.68	6.12	9.63	8.39	2.93	16.26	1.18	15.6

Bacon No Ventilation	27.3	7.10	11.8	15.2	1.69	9.60	14.1	22.6
Bacon Mechanical Ventilation	17.9	4.00	21.5	11.3	0.33	13.0	24.5	26.3
Bacon Natural Ventilation	20.1	5.12	20.7	9.08	0.74	17.1	27.3	18.4
Mean	10.3	5.26	12.2	11.2	2.66	12.0	9.70	16.2

Table S18: The peak concentration (PKC) or maximum concentration metric, which is taken as a measure of the significance of pollutant source or source strength, far from source in other room (upstairs) to 3 s.f. (x 10^5 particles/cm^3).

	H1	H2	H3	H4	H5	H6	H7	H8
Toast No Ventilation	3.34	0.49	0.75	2.93	121	0.80	2.56	0.32
Toast Natural Ventilation	4.56	0.28	0.84	1.59	1.48	0.51	1.62	1.17
Eggs No Ventilation	1.24	1.75	1.97	2.70	0.57	N/A	1.07	1.48
Eggs Mechanical Ventilation	0.42	0.37	0.19	4.85	0.54	1.40	0.64	0.83
Eggs Natural Ventilation	1.15	1.07	2.25	3.02	3.00	1.41	0.79	1.53
Bacon No Ventilation	2.75	0.48	1.22	1.72	0.78	2.04	2.44	2.12
Bacon Mechanical Ventilation	3.39	0.49	1.61	2.16	0.34	3.23	1.40	1.83
Bacon Natural Ventilation	4.43	0.68	1.89	1.56	1.43	4.15	1.68	4.05
Mean	2.66	0.701	1.34	2.57	16.1	1.69	1.53	1.67

Appendix A3: Correlations between Kitchen Volume and Metrics

Figure S1: Correlation of Time to Background (TTB) metric for toasting activities across all eight houses monitored in Chapter 4 vs kitchen volume.

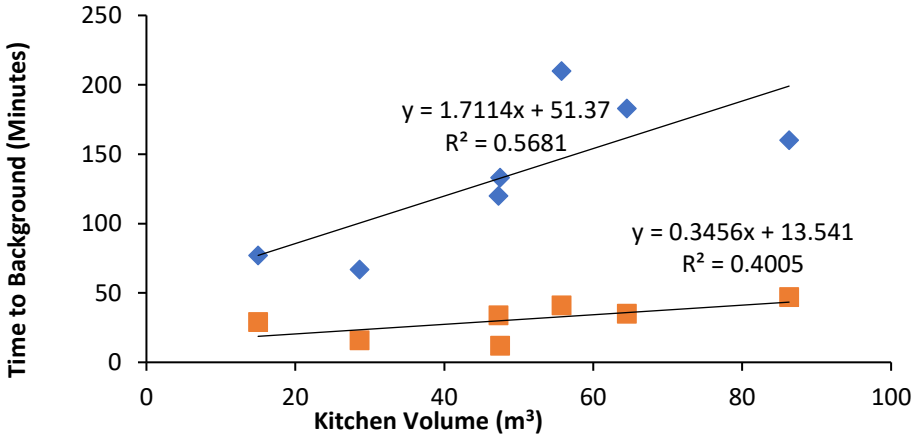


Figure S2: Correlation of Rate of Decay (RTE) metric for frying activities across all eight houses monitored in Chapter 4 vs kitchen volume.

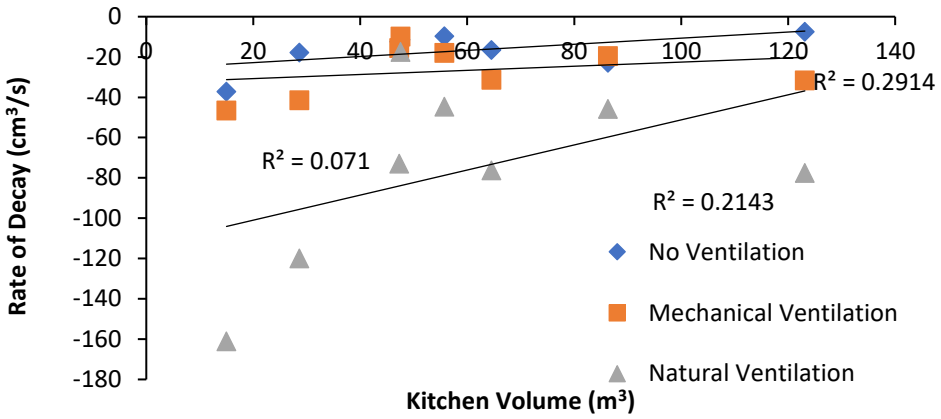
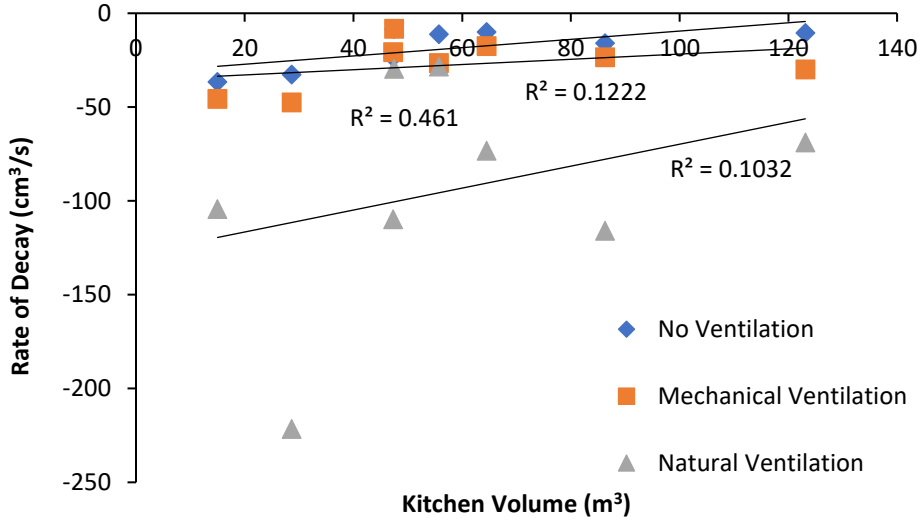


Figure S3: Correlation of Rate of Decay (RTE) metric from oven cooking activities across all eight houses monitored in Chapter 4 vs kitchen volume.



Appendix B: Initial and Full Decay Rates

Table S19: Decay rates (RTE); decay rates for the full duration of decay, for the first 20 minutes (initial decay rate), for the duration of ventilation (corresponding to scenario) and for the period after ventilation, for toasting activities replicated in the Salford Energy House. Rates given to 3 significant figures.

A	Full Duration	First 20 Minutes	Ventilation Duration	After Ventilation
Windows	31.5	62.6	31.5	31.5
No Ventilation	22.5	29.1	22.5	22.5
Windows [20]	15.5	62.5	62.5	15.9
Windows [10]	22.7	72.6	62.1	18.0
Windows [5]	21.9	53.5	12.5	20.7

Table S20: Decay rates (RTE); decay rates for the full duration of decay, for the first 20 minutes (initial decay rate), for the duration of ventilation (corresponding to scenario) and for the period after ventilation, for frying activities replicated in the Salford Energy House. Rates reported to 3 significant figures.

B	Full Duration	First 20 Minutes	Ventilation Duration	After Ventilation
Windows	60.9	72.2	60.9	60.9
Mechanical Vent	83.9	84.8	82.9	82.9
No Ventilation	33.6	47.2	33.6	33.6
Windows [20]	81.8	88.4	88.3	16.1
Windows [10]	58.1	78.9	84.3	31.0
Windows [5]	45.5	71.1	39.5	28.8

Table S21: Decay rates (RTE); decay rates for the full duration of decay, for the first 20 minutes (initial decay rate), for the duration of ventilation (corresponding to scenario) and for the period after ventilation, for repeated toasting activities replicated in the Salford Energy House. Rates reported to 3 significant figures.

C	Full Duration	First 20 Minutes
Windows	62.3	125.1
Windows [20]	20.1	118.7
Windows [5]	32.8	105.7

Table S22: Decay rates (RTE); decay rates for the full duration of decay, for the first 20 minutes (initial decay rate), for the duration of ventilation (corresponding to scenario) and for the period after ventilation, for repeated frying activities replicated in the Salford Energy House. Rates reported to 3 significant figures.

D	Full Duration	First 20 Minutes
Windows	51.6	69.8
No Ventilation	28.1	47.8
Windows [20]	28.1	60.2
Windows [5]	29.1	50.6

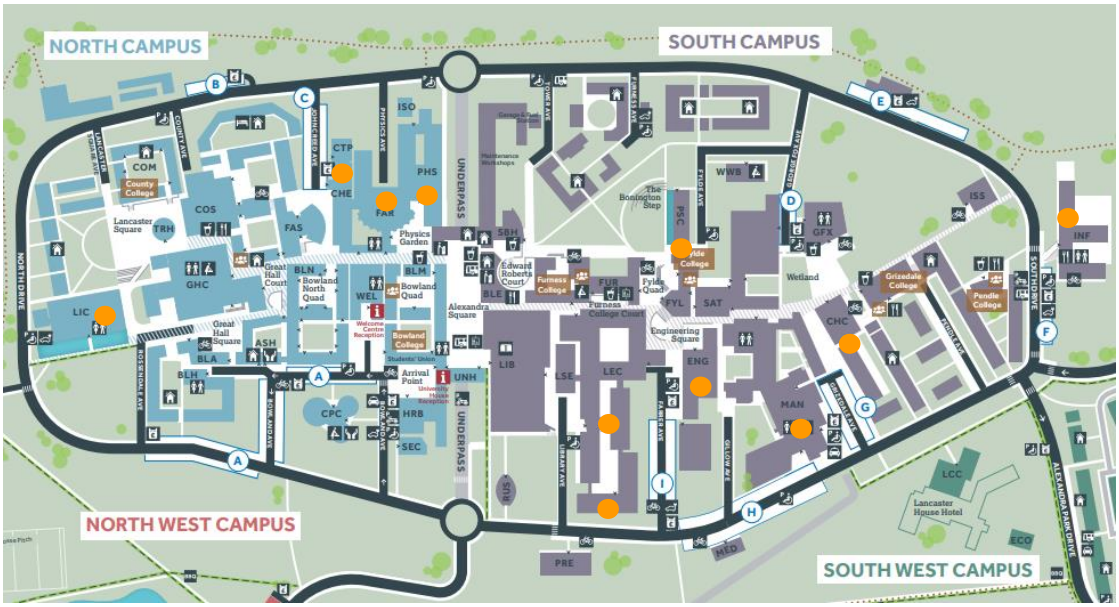
Table S23: Decay rates (RTE) for PNC attributed to toasting activities in the Salford Energy House in the kitchen, living room and upstairs for various ventilation scenarios. Rates reported to 3 significant figures.

	Kitchen	Living Room	Upstairs

No Ventilation	17.4	16.6	25.0
Windows [20]	10.5	20.0	19.4
Windows [5]	12.8	12.8	63.2
Windows	34.1	29.3	23.3

Appendix C1

Figure S4: Lancaster University campus map highlighting locations of BREEAM and Non-BREEAM certified campus departmental and other buildings that were investigated over the course of this study (PHS – Physics, CHE – Chemistry, FAR – Faraday, PSC – Postgraduate Statistics Centre, INF – Infolab, CHC - Charles Carter, MAN – Management School, LEC – Lancaster Environment Centre, LIC – LICA)



Supplementary Information: Raw Data (Semi-Quantification – Top 10 Most Prevalent Compounds) – Full Spectrum – Concentrations given in µg/m^3

Table S24: Full spectrum of semi-quantified VOCs (top 10 most prevalent VOC compounds in each sample) for first round of sampling in Chemistry building (Survey 1).

	PhD [2]	RL [2]	TL [2]	RL [2]	PhD [3]	RL [3]	RL [3]	TL [3]	Corridor [2]	Blank
Isopropanol	5.9	1		1.2						
Acetic Acid	1.8	1.1	1	1.7						
Hexanal	1.4									
Trans-Carene	1.7	1.3	0.9	0.9			2.5			110
2-Ethyl-1-hexanol	9	3.5	1.5	3.3						

Benzyl Alcohol	6									
Nonanol	1.5	0.9		0.8						
Isomenthol	2.2	2.6	1.9	9.7			5.6			
2-Phenoxy-ethanol	5.8	0.9	1.6	1						
Tetradecane	2.5	3.6	2.4	2.9		31				
Acetone		2.6	21	4.2	15	100	48			
Unidentified Containing 'O' circa C13		1.3								
Ethanol			0.8							
Decanal			0.8							
Branched alkane circa C14			1							
para-3-Methene				1.1						
Methyl formate					28					
Formic acid					390					
2,3-Butanedione					33					
Hydroxyacetone					35					
Glycidol					10					
Acetol Acetate					4.2					
Glycerol					>30000	19000	110	4800		55
1-Monoacetin					3600			220		
Menthol					>49000	2400	550	>11000	5000	1900
Hexane						380	310			
Ethyl Acetate						33	85			
Unidentified branched alcohol circa C5						370				
4-Ethyl-3-hexanol						80				
para-3-Methene						22		1900	1800	31
Butyl levulinate						30				
Ethyl ether							4.4			
2-Methyl pentane							6.9			
3-Methyl pentane							6			
1-Methyl-3-(2-methyl-2-propenyl)-cyclopentane								340	230	9
1-Methyl-3-(2-methyl-1-propenyl)-cyclopentane								540	370	18
Cyclohexyl-1-butanol								76		
Isopulegol								130		
para-Menthan-3-one								140		
Menthylchloroformate								79		
para-8-methene									110	
ortho-Menth-8-ene									67	
para-1-Methene									170	7.4
Carane									240	8
1-Cyclohexyl-1-butanol									68	
para-Menthan-3-one									77	
para-Cymene										3.9
Menthylchloroformate										7.3

Table S25: Full spectrum of semi-quantified VOCs (top 10 most prevalent VOC compounds in each sample) for first round of sampling in Chemistry building (Survey 2).

	PhD [3]	**RL [3]**	**RL [3]**	**Corridor [2]**	**TL [2]**	**TL [3]**	**Corridor [3]**	**RL [2]**	**PhD [2]**	**SR [2]**
1-Butoxy-2-Propanol	2.7									11
1-Methoxy-2-Propanol	3.1									8.2
2,2-Dimethylbutane		15								
2-Ethyl-1-hexanol	2.4				3.6			1	9.1	3.6
2-Methylpentane		111	12	2.9					3.3	6

2-Phenoxyethanol					3.8				4.2	
3-Methylpentane		49								3.3
Acetic acid					2.9	2.4	2.7			4.7
Acetone	7.3	102	21	4.7	6.4	2.7	2.9		8	8.4
Benzyl alcohol					3.6					
Branched alkane circa C14								2.9		
Branched alkane containing o circa C11									4.9	
Branched alkane containing O circa C12	2.4			2.7			2.4		2.9	3.6
Branched benzene circa C10			10	3.3						
Branched cyclic alcohol circa C10	51									
Branched cycloalkane circa C10	8.9		13							
Branched cycloalkene circa C10	2.9						2.7		6	
Dichloromethane		44	49							
Ethyl acetate		140	867	2.2		3.8				
Hexane		356	53	2.9					2.4	4.7
Methyl acetate	2.4								2.2	
Nonanal										3.1
Nonanoic acid					4.7	2.9				
Octanoic acid					2.4	2.2				
Tetradecane	3.1			2.7	3.8		2.9	2.9	2.7	
Tetrahydrofuran		91	1022							
Trichloromethane		84	22							
Trichloromethane-d		93	6.4			2.4				

Table S26: Full spectrum of semi-quantified VOCs (top 10 most prevalent VOC compounds in each sample) for concurrent samples taken from inside and outside the laboratory in the chemistry building (Survey 3).

	1A - OUT	**1B – IN**	**2A - OUT**	**2B - IN**	**3A - OUT**	**3B – IN**	**4A - OUT**	**4B - IN**	**5A - OUT**	**5B - IN**
1-Butoxy-2-propanol	2.4				3.1					
1-Methoxy-2-propanol	2.9		2.9		3.1					
2-Ethyl-1-hexanol		5.3		4.2	2.2	3.1		6.2		4
2-Methylpentane	2.4	2.9		2.2		3.6				4.7
2-Phenoxyethanol		7.8		12				4.4		5.3
3-carene	2.4		2.4							
Acetaldehyde		4.9								
Acetic acid			3.8				3.8	2.2	3.1	
Acetone	3.6	4.4	4.2	3.3	8.2	102	4.9	21	3.1	4.2
Decanal	2.4			2.4	3.3	2.4				2.9
Ethanol		6.4		3.3		2.4		2.4		
Hexane	4.4	7.3	4.7	7.6	7.3	8.2	7.6	6.2	5.1	7.1
Isomenthol		5.3								
Methyl acetate	2.7				2.7					
Nonanal	3.6	2.7	4	2.9	4		2.9	2.4		2.7
p-Cymene				4						
p-Menthan-1-ol		71					2.4		2.4	
Tetradecane									3.6	3.1
trans-carene				3.1						