Urbanisation and soil sealing: the effects on ecosystem services and soil carbon

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Declaration

I declare that the work produced for this thesis is my own and has not been previously presented to obtain any other degree. Collaborations with other researchers are properly acknowledged.

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Statement of Authorship

This thesis is prepared in the alternative format as a series of four papers, two of which are published in peer reviewed journals and are presented as the final accepted copy prior to journal editing. The other two papers are intended for submission to journals with the exception of the consolidated bibliography at the end of the thesis, and instances where the papers refer to previous chapters. All papers have multiple authors. Their contribution to each paper is set out below and has been approved by my supervisors. Chapters 1 and 6 present an introduction and discussion to the thesis respectively, and these are not intended for submission.

Chapter 2 is published as:

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RO carried out the literature search, data analysis, co-occurrence analysis of key terms, and prepared the manuscript. JD gave advice on the concept, literature analysis and structure, and contributed significantly to revisions of the manuscript. CS, JNQ and CB contributed significantly to revisions of the manuscript.

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Abstract

Urban soils are increasingly being recognised for the ecosystem services they provide, including soil carbon storage. As urban populations grow, soil ecosystem services in urban areas will become increasingly important. At present, there is a lack of knowledge on the range of ecosystem services provided by urban soils compared to those provided by non-urban soils, and a broad understanding of their provision is lacking. There is increasing interest in the ecosystem service of soil carbon storage as studies have illustrated the ability of urban soils to store large amounts of carbon. In urban areas, soils are affected by urbanisation in numerous ways, including soil sealing with impervious surfaces and the addition of anthropogenic materials, such as construction rubble and waste. At present, our understanding of the effects of soil sealing and anthropogenic materials on soil carbon storage is limited. This thesis seeks to addresses these knowledge gaps by furthering our understanding of urban soil ecosystem services, with a focus on soil carbon storage in sealed soils. It presents a systematic review of urban soil ecosystem service literature followed by a survey of sealed and greenspace soils from across Manchester, UK, to investigate the effects of sealing on soil carbon. The systematic review found that supporting processes and regulating services were most commonly studied, though the multifunctionality of urban soil was being missed in research. The urban soil survey revealed that sealed soils were not always depleted of carbon and, in some cases, legacy carbon stores were present due to black carbon additions which led to carbon stocks that were comparable to, or greater than, greenspace soils. Analysis of functional soil organic matter pools suggested that the legacy carbon store did not contribute to microbially-derived mineral-associated organic matter, indicating that it did not contribute to the persistent organic carbon pool with long residence times. Analysis of deeper soils under sealed surfaces illustrated that soil history was a major controlling factor on soil carbon rather than depth, and it highlighted the importance of heterogeneity in urban soils. This thesis provides new insights into the small but growing body of work on urban and sealed soils. It contributes to our understanding of urban soil ecosystem services,

carbon storage in sealed soils, and the influence of anthropogenic additions and soil history on soil functions. The findings highlight the need to include urban and sealed soil information in soil mapping, planning and construction in urban areas, and to inform best practice when managing urban soils for soil functions and ecosystem services.

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1. Introduction

Soil research has primary developed in agricultural or natural or semi-natural contexts, and, therefore, our understanding of urban soils is relatively limited (De Kimpe and Morel, 2000). Interest in urban soil has increased in recent years in response to the growing awareness of its ability to provide crucial ecosystem services (ESs) in cities (Lehmann and Stahr, 2007; Pavao-Zuckerman, 2012; Morel et al., 2015; Yang and Zhang, 2015; Lal and Stewart, 2017; Vasenev et al., 2018; Calzolari et al., 2020). As over half the global population currently lives in cities, and this is projected to rise to almost 70% by 2050 (United Nations, 2019) there is great importance in understanding urban soil functioning. The ESs underpinned by urban soils are of particular importance for supporting urban resilience and the wellbeing of the urban population (Gomez-Baggethun et al., 2013; Haase et al., 2014; McPhearson et al., 2015). At the local scale, these urban soil ESs include flood mitigation, urban food growing, support for green infrastructure for physical and mental health, capturing air pollution and surface contamination, and physical support for infrastructure. At the local and global scale, urban soils provide nutrient cycling and carbon (C) storage which contribute to climate change mitigation. Many studies focus on only a small number of ESs and there remains a lack of broad understanding and clarity on which ESs have been studied and what we know about their provision in urban areas.

There has been growing interest in the ES of soil C storage as numerous studies have illustrated large urban soil C stores which contribute to climate regulation (Pouyat et al., 2006; Raciti et al., 2011; Edmondson et al., 2014a; Lorenz and Lal, 2015; Vasenev and Kuzyakov, 2018). There has also been interest in nutrient cycling in urban soils as C and nitrogen (N) dynamics are tightly coupled and N plays a key role in soil C sequestration (Lorenz and Lal, 2009; Setala et al., 2016; Averill and Waring, 2018; Trammell et al., 2020; Rocci et al., 2021); while phosphorus (P) has been studied much less in urban soil despite its important role in water quality and plant productivity (Tong and Chen, 2002; Powers et al., 2016). However, most research into urban soil C and nutrients has focused on soil in greenspaces or residential lawns and there is little knowledge on soils under sealed impervious surfaces such as roads and pavements. The extent of sealing varies across cities, but it is estimated that sealed surfaces occupy more than 50% of a city's area (EEA, 2006; Fuller and Gaston, 2009). The rate of soil sealing in Europe was estimated at 1,000 km^2 per year, which is 275 hectares per day or 11 hectares an hour (Prokop et al., 2011). The effect of soil sealing is not extensively studied, though it is considered to prevent the soil from interacting with the wider ecosystem (Scalenghe and Marsan, 2009), impairing soil functions such as water infiltration and run-off regulation (Haase, 2009) and preventing gas exchange (Bardgett, 2016).

We are still lacking a clear understanding of the impacts of sealing on soil C and what the mechanisms of C storage are under sealed surfaces. Studies of sealed soil find that sealing generally tends to deplete soil C and N stores (Raciti et al., 2012; Wei et al., 2014a; Wei et al., 2014b; Piotrowska-Długosz and Charzyński, 2015; Vasenev and Kuzyakov, 2018; Pereira et al., 2021). Sealed soil C stocks have been found to be depleted by 62 – 68 % in Alabama and New York (USA), and Yixing (China) when compared to C stocks in greenspace soils (Raciti et al., 2012; Wei et al., 2014b; Majidzadeh et al., 2017); a loss that is attributed to the removal of topsoil during construction and the lack of organic matter (OM) inputs from plants. However, alternative studies have illustrated that soil C can be similar between sealed and unsealed soils at equivalent depths, suggesting that C losses are greater near the surface due to topsoil loss (Edmondson et al., 2012; Cambou et al., 2018).

We also have limited knowledge on whether the soil C that remains under sealed surfaces is stabilised and will persist with long residence times. It has been suggested that sealing may isolate soil C and reduce loss through decomposition (Vasenev and Kuzyakov, 2018). However, few studies have investigated the stability of the soil C or the biogeochemical cycling in sealed soils, though it has been suggested that organic C (OC) in sealed soils has a lower turnover rate (Wei et al., 2014b), and that water-extractable OM content is much lower in sealed soils (Wang et al., 2021). To date, the vulnerability of sealed soil C to decomposition is yet to be investigated. In addition, the distribution of soil C down the soil profile is also poorly understood in urban soils. Studies of natural or agricultural soils indicate that deep soils play an important role in C storage and that large quantities of C are stored across the whole soil profile (Batjes, 1996; Jobbágy and Jackson, 2000; Salomé et al., 2010; Rumpel and Kögel-Knabner, 2011). In sealed soils, C storage over depth has rarely been considered, though a small number of studies have indicated that deep sealed soils may provide large C stores (Cambou et al., 2018; Bae and Ryu, 2020). As deep urban soils are so often dug up, altered and removed during construction projects, it is important that we understand the functions of deep urban soils and the impacts of urbanisation on them.

In addition to sealing, the heterogeneity that arises in urban soils due to human activity must be considered. Urban soils often contain artefacts, human-made objects that are derived through human activities, such as brick, wood, concrete, metal and plastic (Bullock and Gregory, 1991; Lehmann and Stahr, 2007). The addition of these artefacts leads to the creation of Technosols, defined as either containing large amounts of artefacts, having an impermeable membrane, or a hard material at the soil surface such as stone, asphalt or concrete (FAO, 2015). The accumulation of materials and artefacts over many decades, or centuries, of human settlement can lead to the creation of 'cultural layers' in urban soil, which are known for their high and variable C content (Alexandrovskaya and Alexandrovskiy, 2000; Vasenev et al., 2013; Mazurek et al., 2016). As a result, urban soils are highly complex, and the influence of anthropogenic artefacts needs to be taken into account when understanding soil properties and functions.

Therefore, there are gaps in our knowledge at both the broad scale of urban soil functioning and ES provision, and on a smaller scale in terms of the impacts of sealing, influence of artefacts, and the long-term effects on soil C stores. Addressing these gaps would benefit our understanding of urban soil C, contribute to soil mapping, planning and development for soils in urban areas, as well as informing best practice to enable urban soils to provide important functions and ESs in cities.

1.1 Thesis aims and objectives

This thesis investigates the effect of urbanisation and soil sealing on soil ecosystem services and soil carbon. It explores several knowledge gaps at different scales, firstly, the broad understanding of urban soil-mediated ecosystem services, and secondly, the effects of soil sealing on soil C and nutrients. Thus, this thesis aims to (1) improve our understanding of urban soil's role in providing ecosystem services; and (2) investigate the effects of soil sealing on the ecosystem services of soil C and nutrient storage.

To meet these aims, the thesis addresses the following objectives:

1. To review the literature on ESs provided by urban soils to build a picture of the current knowledge base and identify research gaps (chapter 2);
2. To investigate the effects of soil sealing on soil C and nutrient storage by developing an urban soils dataset across sealed and greenspace soils (chapter 3);
3. To determine the effect of sealing and anthropogenic additions on soil C persistence using analysis of functional pools of soil organic matter (chapter 4);
4. To investigate the influence of profile depth and soil history on the distribution of C and N in sealed soils with varied development histories (chapter 5).

In doing this, the thesis contributes to the small but growing body of work on urban soil ESs and will help to build a greater understanding of the effects of sealing on soil C and nutrient storage and the influence of anthropogenic artefacts and human activities.

1.2 Thesis structure

The thesis consists of four chapters that are either already published or intended for submission and concludes with a general discussion chapter:

Chapter 2 addresses objective 1 and provides a systematic review of the literature on urban soil ESs to create a broad picture of current research. It addresses the gaps in knowledge into which ESs have been studied and what we know about their provision.

It sets out a bibliometric analysis of the literature, a co-occurrence analysis of key terms to understand the research trends, a literature summary for each individual ES, and identifies knowledge gaps and directions for future research.

Chapter 3 addresses objective 2 by investigating the effect of sealing on soil properties, soil C and nutrients. It tests the hypotheses that sealed soils have (i) lower soil C stocks, (ii) lower soil nutrient stocks, and (iii) altered nutrient dynamics compared to greenspace soils. It does this through a comparative survey of urban sealed and greenspace soils across Manchester, UK. The study also considers how artefacts within sealed soils can affect soil properties and functions, influencing soil C and nutrient stocks, and therefore a classification of the samples according to the extent of artefacts is also included.

Chapter 4 addresses objective 3 by investigating the effect of sealing and anthropogenic additions on soil C persistence using a study of functional pools of soil organic matter. It tests the hypotheses that (i) sealed soils will have less mineral-associated OC and therefore less persistent OC; and (ii) soils with anthropogenic additions will have more mineral-associated OC and therefore more persistent OC in both sealed and greenspace soils. Physical fractionation is used to enable the separation of particulate organic matter (POM) and mineral-associated organic matter (MAOM), and OC and N content are determined within each pool to gain insights into the persistence of soil C. The influence of anthropogenic artefacts is also considered in sealed and greenspace soils and their effect on long-term C persistence is assessed.

Chapter 5 addresses objective 4 and explores the effects of depth and soil history on C storage in deep urban soils under sealed surfaces. It investigates the distribution of C and N down the soil profile and tests the hypothesis that soil C and N stocks will decrease with soil depth. It also explores the influence of soil history and development on soil C and N storage and the overall storage capabilities for a 1 m depth sealed soil profile. This is done for three sealed soil profiles in Manchester and Salford, UK with varied development history.

Chapter 6 provides a general discussion and presents wider findings from across the thesis and suggestions for future research.

2. The ecosystem services of urban soils: A review

O'Riordan, R., Davies, J., Stevens, C., Quinton, J.N., and Boyko, C.

This chapter has been published in Geoderma (O'Riordan et al., 2021b).

Abstract

The expansion of urban areas worldwide is increasing the anthropogenic impacts on soil and the role of urban areas in supporting a sustainable future. Thus, urban soils are becoming more important in the delivery of a broad range of ecosystem services (ESs), including carbon storage and climate regulation, biomass provision for food and water flow regulation, and recreational benefits. In this review, we aim to support the development of this emerging research area and, subsequently support the improved treatment and management of urban soil and ES delivery. We present a systematic review of which ESs have been studied and examine trends in research using a co-occurrence analysis of key terms. We then provide a summary review of current knowledge on ESs and identify the gaps in knowledge. Our review highlights that this is a young, but growing, field of research, with a marked increase in publications since 2014. We found that supporting processes and regulating services were most commonly studied, with 88% and 71% of the papers relating to quantitative studies addressing these, respectively. Cultural, provisioning and water-related ESs were relatively understudied, suggesting key gaps for future research. However, this may be attributable to a disconnection between academic communities rather than a lack of knowledge. Fewer than 20% of quantitative studies addressed more than two ESs simultaneously, leading us to suggest that urban soil multifunctionality is a key area for future research, and highlighting the need to integrate understanding of urban soil ESs across disciplines and professions. In addition to this overarching suggestion, we propose six research gaps and opportunities: further research into biomass provision for food; water-related ESs; and cultural ESs; greater geographical representation; further interconnection between research and practitioner communities; and a focus on the future drivers of soil change in urban environments.

2.1 Introduction

More than half of the world's population currently live in urban areas, defined as areas with a population of 10,000 residents or more (DEFRA, 2017), and this is projected to reach almost 70% by 2050 (United Nations, 2019). As urban populations increase, the ability of the urban environment to provide liveable places and support resilient ecosystems becomes more important (Biggs et al., 2012). This, in addition to the risks to human health posed by climate change and air pollution (Jacob and Winner, 2009; Heaviside et al., 2017; O'Donnell and Thorne, 2020), means that it is ever more crucial that we consider how well urban environments are able to maintain the ecosystem services (ESs), namely the benefits people obtain from ecosystems (Millennium Ecosystem Assessment, 2005) that they currently deliver.

Soils play a fundamental role in providing numerous, vital ESs (Dominati et al., 2010; Adhikari and Hartemink, 2016; Jónsson and Davíðsdóttir, 2016; Greiner et al., 2017), and the importance of soil in providing ESs in urban areas is becoming increasingly recognised within the soil science community (Lehmann and Stahr, 2007; Pavao-Zuckerman, 2008; Lal and Stewart, 2017; Ziter and Turner, 2018; Bray and Wickings, 2019). In this review, we consider urban soils to be all soils located within urban areas. Urban soils are included within SUITMA (Soils of Urban, Industrial, Traffic, Mining and Military Areas), defined as soils strongly modified by human activities with drastic changes in composition and function, though in urban areas, they can include both highly-transformed soils and pseudo-natural soils (Morel et al., 2015). In this review, we limit our focus to soils within urban areas to enable a focus on the provision of ESs in areas where the majority of people live. In urban areas, urban soil underpins many ESs that provide importance for human wellbeing and urban resilience (Gomez-Baggethun et al., 2013; Haase et al., 2014; McPhearson et al., 2015). Locally, these services include flood mitigation, buffering the urban heat island effect, capturing air pollution, physical support for infrastructure, urban food growing and access to greenspace for mental and physical health; whilst at local and global scales, they contribute to nutrient cycling and carbon (C) storage.

Urban soils are able to provide many of the same ESs as non-urban soils (Pavao-Zuckerman, 2012; Morel et al., 2015; Pouyat et al., 2020). At present, however, there is relatively limited knowledge on their quantification as compared to non-urban soil ESs. Much work since the development of The Economics of Ecosystems and Biodiversity (Kumar, 2010) and The Common International Classification of Ecosystem Services (Haines-Young and Potschin, 2018) has placed a focus on ecosystem goods and services that are directly beneficial to humans, allowing the economic valuation and accounting of ES. Whilst this valuation makes the concept useful to policy and decision makers, there remains a need to further understand specifically how urban soil supports ESs. Research into urban soil ESs is still in its infancy and much work is at the level of soil processes, functions or properties. As such, it is necessary to focus on, and distinguish between, supporting processes that drive soil functioning, and soil ESs that are directly beneficial to humans (Dominati et al., 2010; Baveye et al., 2016).

The study of urban soil ESs is slowly gaining momentum, often with a theoretical focus on the potential ESs that can be provided (Morel et al., 2015; Vasenev et al., 2018), or through improving methods of quantification and integration into planning (Blanchart et al., 2018; da Silva et al., 2018). However, there remains a gap in bringing together what is currently known within the research community about urban soil ES provision. There is a need to gain a better understanding of which ESs are provided by urban soils; the extent to which individual ESs have been studied; how they will be altered by future drivers of change such as climate change; and how we can manage urban soils to deliver ES, now and in the future. This review serves to address these needs by bringing together the literature on urban soil ESs to provide an understanding of what we currently know, analysis of the trends in research and an identification of gaps in knowledge.

We firstly present a bibliometric analysis of the urban soil ES literature, analysing which ESs have been most studied, where and at what soil depth; and explore the structure of the research community using a co-occurrence analysis of key terms. We then provide a summary review of knowledge on individual ESs delivered by urban soils, reviewing what has been studied and where the gaps in knowledge are. Lastly, we make suggestions for the direction of future research to aid the understanding of urban soil ESs and optimise their provision.

2.2 Material and methods

2.2.1 Literature search

We performed a literature search to gain an understanding of which urban soil ESs have been most studied and where. There was a focus on the use of ES terminology to identify studies that employed an ES framing. We also included terminology associated with soil processes and functions in addition to ES, as these terms are still used interchangeably within the soil science community (Schwilch et al., 2016), and the ideas of ESs and ecological functions are closely related (Vasenev et al., 2018). This interrelation is recognised by Baveye et al. (2016) who stressed that it is important to consider both soil functions and ecosystem services, so long as they are articulated in relation to soil properties and processes (Bünemann et al., 2018).

A search of English language literature was performed in April 2020 on Web of Science for urban AND soil* in the title, combined with "ecosystem service*" in the topic (title, abstract and keywords). A second search was run for "urban soil*" AND "ecosystem service*" in the topic. A third search was run for ("soil process*" or "soil function*") AND urban* in the topic. These three searches were then combined using the OR operator. The complete search had the following search string:

(TI=(urban AND soil*) AND TS="ecosystem service*") OR (TS=("urban soil*" AND "ecosystem service*")) OR (TS=(("soil process*" OR "soil function*") AND urban*))

The same search was run on Scopus and documents were collated together. Book chapters, meeting abstracts and conference reviews were excluded. An initial review of the documents was undertaken and those without an urban focus were removed, which left 178 papers that were relevant to urban soil and ES.

2.2.2 Data analysis

The literature was first separated into three categories: those that measured ESs through empirical data or modelling studies were referred to as 'quantified' papers; those that only discussed ESs in relation to urban soils were referred to as 'discussion' papers; and those that did not specifically quantify or discuss ESs were referred to as 'general urban soil' papers. Where papers had collected data that provided information about the listed ESs, whether explicitly described as an ES or not, they were classed as 'quantified' papers. Review papers were included in the 'discussion' or 'general urban soil' papers.

We undertook the bibliometric analysis on all categories of literature. We then carried out more detailed analysis on the 'quantified' papers to investigate which ESs had been studied, which were commonly studied together, and which soil depths were most recorded. The findings in these 'quantified' papers were then used to present the summary review of urban soil ESs.

To capture how urban soil supporting processes and ESs are being studied and at which level, the framework of soil ESs proposed by Dominati et al. (2010) was used (Table 2.1). The framework distinguishes between supporting processes that drive soil functioning (such as nutrient cycling, water cycling or soil biological activity), and ESs that are directly beneficial to humans, which include provisioning, regulating and cultural services. The definitions given by Dominati et al. (2010) were used to categorise the supporting processes and ESs identified in the 'quantified' papers, and are provided in the supplementary material (Appendix 1).

Some studies measured both a supporting process (e.g. microbial activity) and a regulating service that is related to the supporting process (e.g. C storage); in these cases, the papers were classed as measuring both. While these processes and services are interlinked, they have been analysed in this way to build an understanding of which supporting processes and ESs have been studied in detail, and in addition, how researchers refer to them and approach studying them.

Table 2.1: The list of soil supporting processes and ecosystem services given in Dominati et al. (2010) used in this manuscript.

Category	**Supporting Process or Ecosystem Service**
Supporting Processes	Nutrient cycling; water cycling; soil biological activity
Provisioning ESs	Food, wood and fibre; physical support; raw materials
Regulating ESs	Flood mitigation; filtering of nutrients; Biological control of pests and diseases; Recycling of wastes and detoxification; Carbon storage and regulation of greenhouse gas (GHG) emissions
Cultural ESs	Spirituality; knowledge; sense of place; aesthetics

2.2.3 Co-occurrence analysis of key terms

The titles and abstracts of the 178 papers collected in the literature search were analysed using the VOSviewer software (Van Eck and Waltman, 2010) to identify the most common terms and co-occurrences between them. A threshold of 5 occurrences of each term was used to identify common terms in the literature (one count per title/abstract rather than all counts of each term). A thesaurus file was used to simplify terms for consistency (such as SOC to soil organic carbon, or soil C to soil carbon). A relevance score was applied by the software that filters out generic terms such as 'method' or 'result', and which helps cluster together topic-specific terms (Van Eck and Waltman, 2011). The co-occurrence network is presented to show terms with the most occurrences, links between them and where clusters form between the terms. The clusters were set to a minimum of 25 terms per cluster to enable themes to be visualised.

2.3 Results and discussion

2.3.1 Analysis of urban soil ES literature

2.3.1.1 Bibliometric analysis of literature

The distribution of the literature with publishing year and geographical scope is illustrated in Figure 2.1. The number of publications on urban soil ESs is relatively small and recent compared to that of soil ESs, with the oldest paper found dating from 1997. Papers studying urban soil ESs did not become more common until 2014, after which the number of publications generally increased, with the most published in 2018 (Fig. 2.1 a).

Much of the literature identified relates to studies in Europe (42%) as shown in Fig. 2.1 b. Following this, 22% of literature was based in the continent of North America. Very few studies were undertaken in Africa, Australia and Oceania or South America (2%, 2% and 1% respectively). Figure 2.1c provides this data at the country level, where it was given, and indicates that most English language research was undertaken in the USA which has nearly twice the number of papers than the next highest publishing countries, China, UK, France and Germany. Many papers do not undertake research at the individual country or continent scale but take a global perspective, these have been labelled as 'World' in Figures 2.1b and 2.1c, which provide examples of review or discussion papers.

The majority of papers (125) were those that had 'quantified' ES, while 32 were classed as 'discussion' papers and 21 were 'general urban soil' papers. In the discussion papers there was a focus on soil biological activity and C storage, however, most discussion papers (47%) mentioned numerous ES categories. Biomass provision for food and cultural services were poorly represented, with food being mentioned in two discussion papers, and cultural services mentioned in only one.

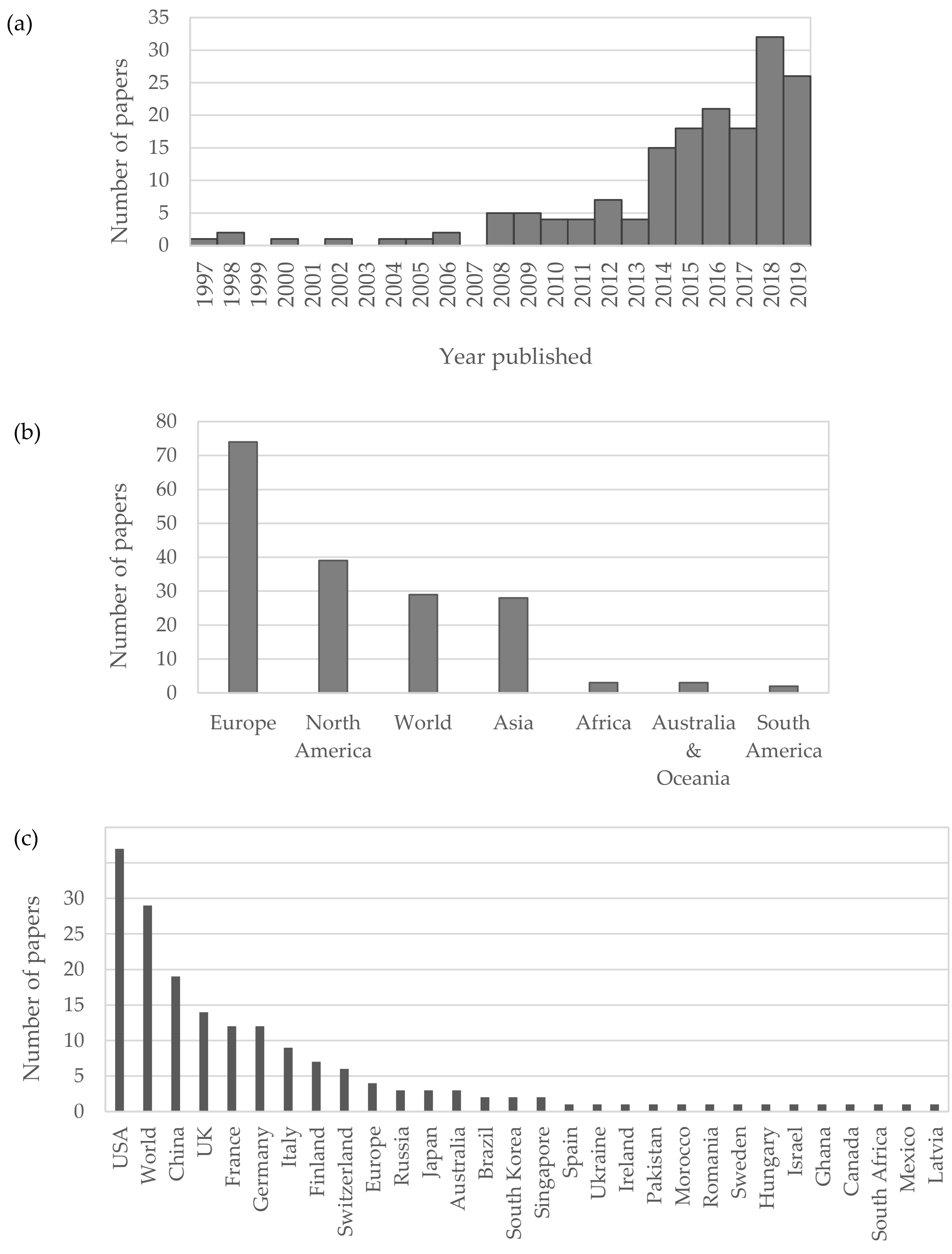

Figure 2.1: (a) Number of papers published between 1997 and 2019 using the search string specified; (b) number of papers published by global region; (c) number of papers published with scopes at different geographical scales: country, continent or global.

2.3.1.2 *Specific ES analysis*

To provide an understanding of which individual ESs had been studied, an analysis of specific ESs was undertaken on the 125 papers that had quantified data, as illustrated in Fig. 2.2. A majority (88%) of these quantified studies focused on supporting processes, with 42% of the studies measuring soil biological activity, 34% measuring nutrient cycling and 12% measuring water cycling. The predominance of studies focusing on these supporting processes, particularly nutrient cycling and soil biological activity, highlights their importance in understanding soil functioning and their support in providing ESs. However, there appears to be less of a focus on water cycling as a supporting process in urban soils. This may be because urban soil water dynamics are commonly studied in relation to water storage capacity or urban water management, and therefore, these papers will be captured within the regulating service of flood mitigation.

Regulating ESs were also frequently studied in the quantitative literature (71% of studies), with 30% measuring C storage and regulation of greenhouse gases (GHG), and 21% measuring the recycling of wastes and detoxification. Flood mitigation appeared in only 9% of quantified papers, with only a small number measuring urban stormwater management as an ES. This does not reflect the extent of research and practical experience within professions working on urban water and sustainable urban drainage systems (SuDS) (Ciria, 2013; Davis and Naumann, 2017; Schifman and Shuster, 2019). It does, however, suggest that stormwater management is commonly seen as a problem to rectify rather than framed as the soil ES of flood mitigation; and as such the knowledge developed in engineering and water disciplines may not be reaching the wider ES community. There was also a lack of studies on the regulating service of biological control of pests and diseases in urban soils.

Provisioning ESs were less often studied, with the service of food, wood and fibre provision making up only 3% of the quantified papers. This is in contrast to research in non-urban soils where food provision is often quantified as one of the most important soil ESs (Adhikari and Hartemink, 2016; Holt et al., 2016). Urban agriculture is a well-established practice across the world, represented by a broad range of literature (Orsini et al., 2013; Ackerman et al., 2014; Mok et al., 2014; Edmondson et al., 2020); however, our findings suggest it is rarely studied in the context of urban soil ESs, and may have been missed from the literature as it does not explicitly mention soil ESs. The later average publication date for food, wood and fibre provision studies (Fig. 2.2) may, however, suggest that it is a growing area for ES studies. Physical support for built infrastructure, such as roads or buildings, occupied only 2% of the quantified papers which does not reflect the communities of research and practice in urban soil geotechnics (Trombetta et al., 2014; Denies et al., 2015; Vardon, 2015; Price et al., 2018). This suggests that while well-established, engineering and geotechnical communities may not be considering urban soils within an ES framing. In addition, the literature search did not identify any studies on raw materials from urban soils, or on the concept of urban mining, the recovery and reuse of resources from waste materials (Arora et al., 2017).

None of the studies undertook survey or analytical work on cultural services from urban soils. There is a large body of work on the cultural and archaeological significance of soils capturing historical and societal information, referred to by some as cultural layers within cities (Burghardt, 1994; Vasenev and Kuzyakov, 2018). However, this work does not appear to use the terminology of ESs, perhaps because ES research has largely been developed by ecologists and economists rather than by heritage researchers (Hølleland et al., 2017). In addition, there is a growing body of evidence for the importance of access to nature and urban greenspaces for both mental and physical health benefits (Pretty et al., 2011; Lovell et al., 2018; Chen et al., 2019); however, these benefits are often captured in relation to trees or urban forests rather than soils. The approach to studying cultural ESs remains an on-going debate (Fish et al., 2016), and as such, their study in both urban and non-urban soils is still relatively rare.

Across all individual ESs quantified, the greatest number of studies were undertaken in the USA, followed by China and European countries. After the USA, a relatively large proportion of the soil biological activity studies were undertaken in France; while for C storage, numerous studies were completed in the USA, UK and China. A small portion of the quantitative literature (6%) focused on Technosols, defined as soils dominated by technical human activity and evidenced by a substantial presence of artefacts or an impermeable constructed geomembrane (Rossiter, 2007). These papers focused mostly on constructed Technosols and their effects on soil biological activity, water infiltration and nutrient cycling, and were almost exclusively undertaken in France.

2.3.1.3 Interrelation between ESs studied

Most papers (57%) studied only one ES, while 26% studied two services, 15% studied three, 2% studied four and only 1% studied five. Where more than one service was studied, common pairings of services were quantified together which illustrated the interrelation between them. There was a predominance of supporting processes being studied together, for example, 48% of nutrient cycling papers also measured soil biological activity, two processes that are particularly intertwined (Bardgett, 2005); and 47% of water cycling papers also measured nutrient cycling, highlighting these measures as important indicators for urban soil functioning.

Instances of regulating services studied together were less common, for example, there were only four papers where flood mitigation was studied alongside C storage. Only two papers studied filtering of nutrients alongside recycling of waste and detoxification, suggesting the link is not being made between the pools of contaminants and the ability of soil to filter these or prevent their release into the environment.

The interrelation between supporting processes and regulating or provisioning ESs varied. Of the papers that measured C storage, 39% also measured nutrient cycling; while only 21% measured soil biological activity, suggesting only a small number of papers are undertaking work on the connection between soil biota and C storage in urban soils. Water cycling was not commonly measured with flood mitigation suggesting these services are thought of separately and by different groups of researchers. In addition, nutrient cycling was rarely measured alongside food provision, which again suggests different groups of researchers or practitioners each with their own terminology and data collection methods.

The lack of interrelation across service types highlights that supporting processes and ESs are not commonly considered together; and that regulating services are not often studied together or with provisioning services. This lack of studies on multiple ESs illustrates that the opportunity to quantify the multifunctionality of soil is being missed. There is a need to measure supporting processes to understand the basis of ES provision, but there is also a need to quantify regulating and provisioning services together to allow the multifunctionality of soil to be included in urban planning and decision making.

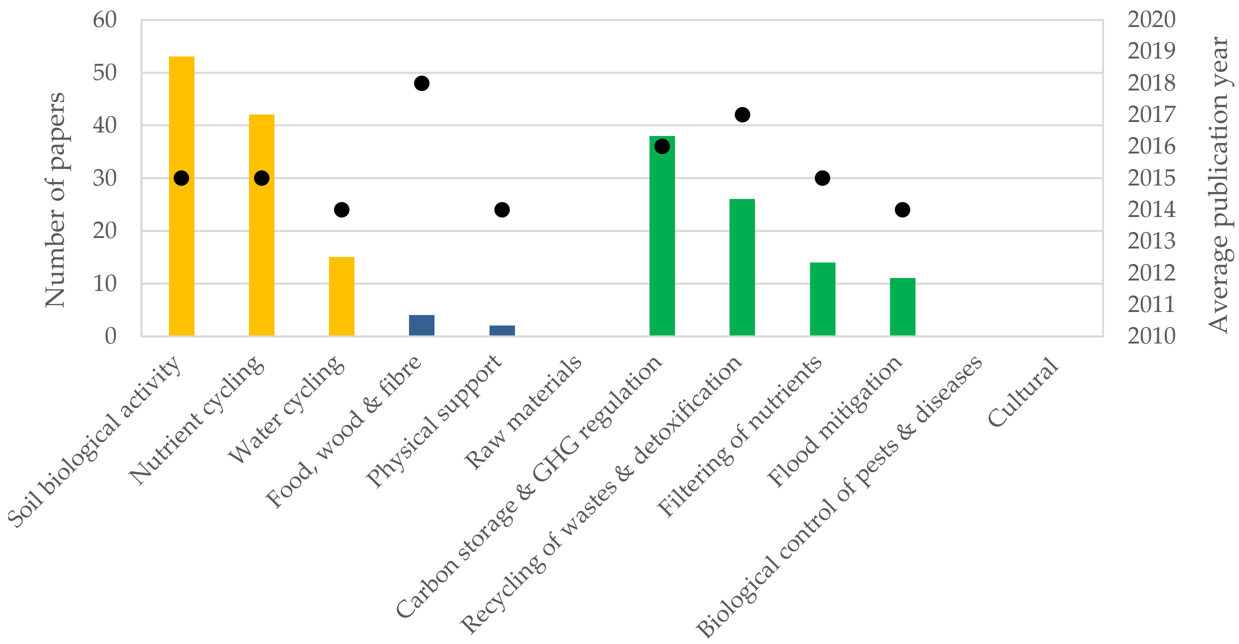

Figure 2.2: Number of papers measuring supporting processes and ESs. Papers included are those that quantified ESs (number=125). Yellow, blue and green columns represent supporting processes, provisioning and regulating ESs respectively. Circles indicate average (mean) publication year for each ES, shown on secondary axis.

2.3.1.4 *Depth of urban soil studied*

Data on the maximum depth and number of measurements down the soil profile was gathered from the literature, where it was provided (Fig. 2.3). Of the 104 papers that gave depth information, the majority of papers studied soil between 0-20 cm (63%), while 14% studied down to 40 cm, and 12% studied down to 100 cm. Papers studying deeper than 100 cm (5%) were restricted to those that used deep cores to study subsoil drainage (Herrmann et al., 2017), lysimeters to observe leachate (Cannavo et al., 2018; Yilmaz et al., 2019), soil chemistry under sealed surfaces (Kida and Kawahigashi, 2015), and risks associated with soil swelling (Vallone et al., 2008). Most papers studied just one soil depth (70%) while a smaller number of papers investigated differences between two, three or more than three depths (10%, 15% and 5% respectively).

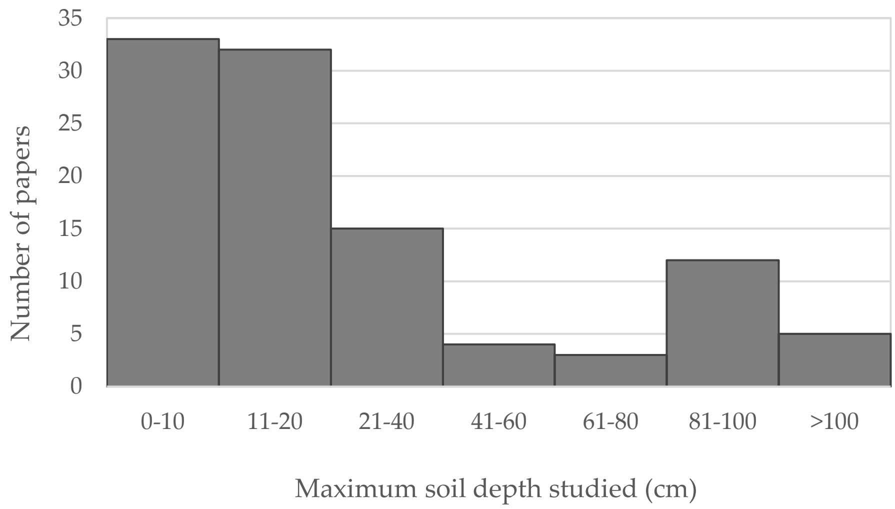

Figure 2.3: Maximum depth (cm) at which papers studied urban soil ESs.

2.3.1.5 *Co-occurrence analysis of key terms*

An analysis of the co-occurrence of key terms in the literature led to a network visualisation of the terms and links between them. Three clusters of terms are identified, highlighted by colour in Fig. 2.4, with clusters representing similar observations or processes occurring together in the literature.

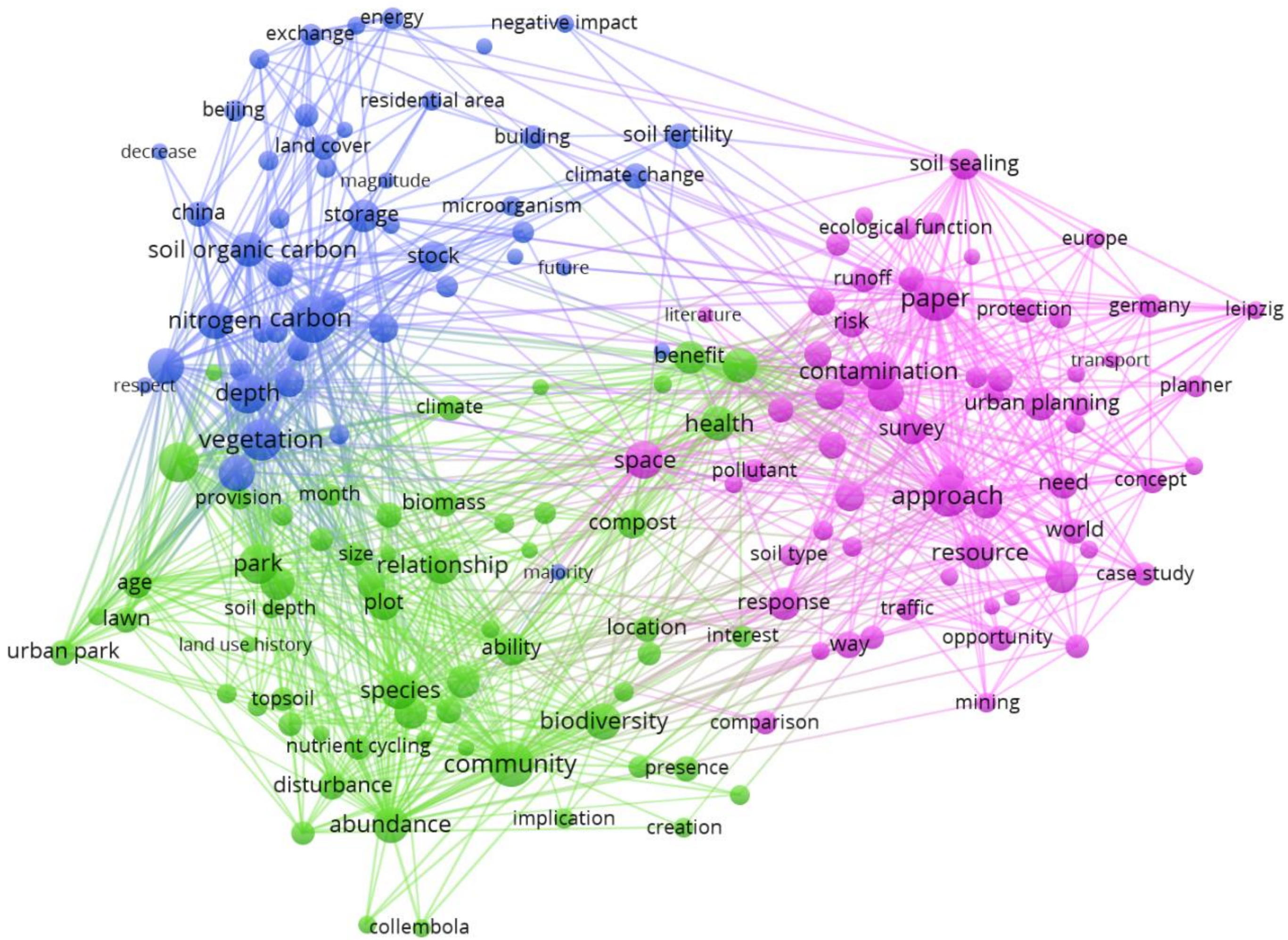

Figure 2.4: Co-occurrence analysis of key terms within the urban soil ecosystem service literature. Nodes represent terms that occur at least five times, with the size of node denoting the number of occurrences. Vertices and relative distance of nodes illustrate the co-occurrence of terms. Three clusters where the interconnections of terms are strongest are identified, denoted by colour: C and nutrients (blue); soil biodiversity (green); and the challenge of urban soils (purple).

Within the C and nutrients cluster (blue) there is a focus on stocks of C and nutrients and the impacts of urban land cover on their storage, such as soils under buildings or impervious surfaces, or different vegetation types such as urban forests or lawns. The soil biodiversity cluster (green) highlights a separate group that focuses on the abundance and diversity of species, their distribution across different green infrastructure types, their activities such as nutrient cycling, and the consequences of urbanisation and disturbance on them. Finally, there is a third cluster focused on the challenge of urban soils (purple), which includes the impacts of urbanisation, risks to soil such as soil sealing, excess runoff and contamination, opportunities to manage and plan to protect urban soil better, and strategies to highlight its importance in planning documents.

The clusters of key terms reflect what is shown in the specific ESs analysis (section 2.3.1.2), that research tends to focus on supporting processes with a predominance on soil biological activity, as well as soil C and nutrient stocks. There is an area of cross over between the blue and green clusters where terms represent a range of green infrastructure types that have been studied such as urban parks, lawns and different vegetation types. These terms co-occur together and are relevant for both the C and nutrient cluster and the soil biodiversity cluster. This aligns with patterns found in the ES literature analysis (Fig. 2.2), which showed that most studies focused on soil biological activity, nutrient cycling and C storage.

There is a distinct lack of terms associated with water across the co-occurrence analysis, be that flooding, water holding or water cycling, and while the terms soil sealing and impervious surface are included, they are not connected to issues of flooding. However, the ESs literature analysis (section 2.3.2) illustrates a small but important number of studies that investigate water cycling, runoff and flood mitigation. These studies use a range of measurements of soil water such as percolation, infiltration, water holding, runoff, saturated / unsaturated hydraulic conductivity and field capacity, and therefore, it is possible that these terms do not appear frequently enough in the literature to be captured in the co-occurrence analysis.

Another notable gap in key terms are those that relate to food and urban growing which correlates with the lack of literature on food provision in the ES literature analysis, reiterating the lack of food provision terminology used in the urban soil ES community. Likewise, cultural services were also not represented within the co-occurrence analysis, representing the lack of studies found in the ES literature.

2.3.2 *Summary review of urban soil ESs*

Having analysed which urban soil ESs have been quantified, where this was undertaken and the nature of the research community in 2.3.1, here we provide an overview of research reported in the 'quantified' literature identified by ES category. We prioritise primary research studies in order to provide some insight into what is known and where future research gaps may lie.

2.3.2.1 *Supporting processes*

Nutrient cycling

Human activities and land use have the potential to alter nutrient cycling in urban soils due to direct and indirect additions and removals of nutrients, and modifications to factors affecting nutrient cycling.

Several studies have found that soils under some urban land uses can have high nutrient contents. Schindelbeck et al. (2008) compared land use in New York state and Baltimore, finding that soils from a recreational park and brownfield plot had higher organic matter content and mineralizable nitrogen (N) content than soil from a non-urban vegetable farm. In Lahti (Finland), soil in a managed garden site showed consistently higher nutrient content compared with human-made soil on a landfill site (Vauramo and Setala, 2010). In Leicester (UK), allotment soils had higher amounts of organic N than soils from surrounding intensive arable fields, which was attributed to additions of compost or manure (Edmondson et al., 2014a). The time period over which the soil has been under a particular land use is also an important determinant of nutrient status. Soil organic matter and nutrient contents have been found to correlate with park age (Setala et al., 2016) and housing age (Cobley et al., 2018) in studies in Finland and the USA respectively.

Conversely, some urban land uses and conditions led to a reduction in nutrient contents. For example, Herrmann et al. (2017) found that imported soil, used to fill excavations on previously developed land, showed less nutrient support for plant growth with lower N levels than pre-existing soils at the site. Nutrients have also been found to be depleted in areas where an accumulation of heavy metals was apparent (Zhao et al., 2013).

Phosphorus has been studied significantly less compared to other macronutrients in urban soils; however, it is likely that it would be equally altered by urbanisation through physical modifications such as land use, vegetation types in greenspaces (Setala et al., 2017), human or industrial waste additions (Yang and Zhang, 2015), and altered soil biology such as earthworm activity (Amosse et al., 2015). Likewise, there were few studies that considered other physical modifications to the urban environment and their effects on nutrient cycling, for example, connections were not commonly made between altered urban hydrology, microclimate, aeration and soil structure and how these might affect urban soil nutrient cycling.

Water cycling

The primary factors influencing water cycling in urban areas are the extent of impermeable surfaces, soil infiltration capability and drainage, and evapotranspiration (McGrane, 2016). However, other factors also contribute to altered soil water cycling, including the heterogeneity of urban soil, greenspace management, altered horizons and compaction due to construction activities.

A number of studies identified by the literature focused on infiltration, soil moisture dynamics and water holding. One of the earliest papers identified mapped infiltration rates for Hannover (Germany), including areas covered by roads and buildings as well as open soils and vegetation covered areas (Bartsch et al., 1997). In a modelling study in Leipzig, Haase (2009) found that water cycling had accelerated due to increased sealing with impervious surfaces, leading to reduced water holding capacity in favour of increased runoff. Recent modelling studies have considered soil moisture dynamics across different world cities with varying levels of permeable surfaces (Revelli and Porporato, 2018), as well as the effects of developments on groundwater recharge and the sensitivity of this to future climate scenarios (Manna et al., 2017).

The link between organic matter and soil water holding, as observed in traditional soil science (Rawls et al., 2003; Minasny and McBratney, 2018), has also been observed in the urban soil literature. A recreational park soil in New York state had higher available water capacity compared with farm or brownfield soils, attributed to the high organic matter content (Schindelbeck et al., 2008); while Oldfield et al. (2014) found that compost additions to soil led to increased water holding capacity in the New York City Afforestation project. In urban gardens in Zurich, Tresch et al. (2019a) found high correlations between C mineralisation and water holding capacity as part of a study on soil multifunctionality. They found that soil moisture and disturbance, driven by watering and tilling, were key drivers in structuring plant and soil fauna communities, which in turn influence multifunctionality, thus highlighting the importance of watering regimes in soil multifunctionality.

While extensive methods are used to measure natural and agricultural soil physical and hydrological properties, measurements of infiltration in urban soil present unique challenges due to the presence of artefacts (Rhea et al., 2014). There are a limited number of studies into the properties of Technosols in relation to water cycling, and methods to investigate hydraulic properties of several Technosols were compared by Yilmaz et al. (2019); while soil water in Technosols made with waste were studied by Cannavo et al. (2018) who found that physical properties were not necessarily a limitation to tree growth.

Soil biological activity

A recent review by Guilland et al. (2018) found that studies on the biology of urban soils made up around 2-3% of all studies of soil biology. Whilst this is in line with the extent of urban land cover globally, arguably a greater focus on urban soils is needed as there is a clear relationship between biodiversity, ecological processes and ES provision (Mace et al., 2012), and this is closely linked to the location of the majority of the population. Guilland et al. (2018) found that most studies were about microorganisms, nematodes and arthropods (33%, 28% and 21% respectively), and that most studies focused on ecotoxicology or bioaccumulation of contaminants rather than the ecological and functional aspects of soil biological communities.

Contrary to assumptions, soils in urban areas do not always have compromised soil fauna. Based on a study of microarthropod biodiversity, urban soils may provide the same level of biological quality as forests (Joimel et al., 2017); and while not picked up in the ES literature, Ramirez et al. (2014) found that the breadth of microbial diversity in Central Park in New York was similar to microbial diversity across the world. Direct comparisons of urban to non-urban soils can, however, lead to varying conclusions, as urban land uses studies in China and Finland have found lower soil microbial biomass than in natural forests (Zhao et al., 2013; Francini et al., 2018); whereas microbial activities of urban soils in Stuttgart were comparable to agricultural or forests soils (Lorenz and Kandeler, 2006).

A variety of factors have been found to influence soil fauna distribution within urban areas (Santorufo et al., 2014; Xie et al., 2018; Joimel et al., 2019; Tresch et al., 2019b). Soil parameters were found to exert a stronger influence on soil fauna than plant communities in vegetable gardens (Joimel et al., 2019); however, Tresch et al. (2019b) found that plant species richness affected soil fauna diversity and microbial activity in urban gardens. It has also been observed that the typical pattern of plant-microbe associations seen in non-urban soils has also been seen in urban soil, such that urban soil bacterial and fungal communities can respond to plant functional groups (Hui et al., 2017). Nevertheless, there remains limited understanding of what influences microbes' distribution in urban soils (Wang et al., 2018).

Urban land use can also have an effect on soil fauna. Urban soils have been observed to exhibit greater functional diversity than other non-urban land uses, particularly in roadside tree soils in Beijing (Zhao et al., 2013); and greater species diversity in park and roadside soils compared to residential soils in Chicago (Wang et al., 2018). The history of disturbance also has an influence, as the relationship between soil biota and physicochemical variables can vary with soil age (Amosse et al., 2016); and park age can shape composition of microbial communities (Hui et al., 2017).

Microbial activity can be affected by pollutants such as heavy metals and pesticides (Gan and Wickings, 2017). For example, Ivashchenko et al. (2019) found microbial C-availability and organic matter decomposition were lower in industrial and residential zones of Moscow where there were higher levels of heavy metals, and metal contaminated soils have also been shown to have lower levels of nitrifying bacteria and a lack of fungi (Hartley et al., 2008).

2.3.2.2 Regulating services

Flood mitigation

The ability of urban soils to provide flood mitigation is largely influenced by land use and land surface treatment (Haase, 2009; Wheater and Evans, 2009). Urban forest soils have been shown to have better drainage than soils on residential or commercial land (Dobbs et al., 2011), and have higher runoff regulation than other urban land uses (Ziter and Turner, 2018). The size of urban forest patch was not found to affect hydraulic conductivity in a study by Phillips et al. (2019), who conclude that the protection of urban forest patches, whether small or large, can potentially contribute to urban stormwater management.

Sealed land surfaces, such as impervious roads and paving, notably increase surface runoff. Runoff values start to double when impervious surfaces cover >20% of land,

and a model for Leipzig has shown that runoff can reach over 75% of the annual precipitation level when areas are >80% impervious (Haase, 2009). A more recent study shows that runoff values could increase by >20% in highly sealed areas (Ungaro et al., 2014). Where permeable soils remain, for example around the base of street trees, there is an increase in rainfall infiltration (Revelli and Porporato, 2018). One possible solution to increased runoff is the use of suspended pavement systems, such as those above tree pits, which in a study in Knoxville (USA) reduced 99% of measured runoff volumes, and captured runoff from 79% of storms (Tirpak et al., 2019). Inclusion of soil sealing management in planning strategies and policies has also been considered to reduce the growth of sealed areas (Artmann, 2015; Artmann, 2016).

The extent of human disturbance, compaction and addition of anthropogenic material to the soil itself also influences the capacity for flood mitigation. Imported fill soils used in construction are variable, depending on the material used, but some have been shown to have greater infiltration and drainage than pre-existing soils (Herrmann et al., 2017). Soils with compost mixed into the subsoil and tilled had twice the saturated hydraulic conductivity of undisturbed soils, and 6-11 times that of soils subjected to topsoil removal and subsoil compaction (Chen et al., 2014). This suggests that some treatments may have potential in aiding stormwater mitigation.

Filtering of nutrients

Soils can filter and retain numerous organic or inorganic compounds and solutes and prevent them from reaching water courses (Dominati et al., 2010). The ability of soil to act as a filter can be influenced by vegetation cover; however, only a small pool of studies has considered the link between vegetation and urban soil as a filter. Urban soils under tree canopies have been found to have higher C to N ratios than soils under grass due to the higher C to N ratio in tree litter, and thus, are more able to buffer localised N fertilisers or atmospheric N deposition (Livesley et al., 2016). In a study by Ziter and Turner (2018), urban soils in grasslands and open spaces were found to have the lowest available phosphorus (considered as a proxy for potential P runoff) compared with urban forests and developed land in Madison (USA).

In addition to plant influences, other forms of C in urban soil may contribute to water filtration. Black C accumulation in urban soils may act as a sorbent of contaminants, and in combination with sufficient infiltration rates, may lead to improved water filtration and improved water quality in urban greenspaces (Schifman et al., 2018). A possible practice to improve soil filtration is the use of suspended pavement systems, as mentioned in section 2.3.2.2, on which a study has shown the concentration of influent suspended solids to be significantly reduced, demonstrating the bioretention potential of these systems to remove pollutants from urban runoff (Tirpak et al., 2019).

Urban soils can, however, act as a source of nutrients or pollutants when the soil's ability to filter them is compromised, and thus, the retention of pollutants can become an ecosystem disservice. Road salt can leach from urban soils into water courses, with riverine Cl^- loading downstream of Calgary (Canada) attributed to increasing inputs of road salt (Kerr, 2017). Remediation of degraded urban soils often involves additions of compost that can lead to excess nutrient leaching and impacts on urban water quality. In a degraded urban soil experiment, N and P losses were considerable prior to vegetation establishment; however, once vegetation was established, N and P losses reduced to background levels (Basta et al., 2016). To reduce leaching risks, Heyman et al. (2019) identified a range of acceptable compost characteristics that would be beneficial for soil remediation without causing nutrient leaching. As with other land covers, plant type as well as litter inputs and the ratio of soil bacteria to fungi can also influence nutrient leaching. For example, urban soils with labile litter inputs and greater associated soil bacteria have been shown to leach more inorganic N than soils under recalcitrant, less readily decomposable litter, which have greater associated soil fungi (Vauramo and Setala, 2010).

Recycling of wastes and toxins

Soil has the ability to degrade and decompose some waste and chemical contaminants; however, if levels are high and the soil holds onto large amounts, it can represent a source of contamination to people living in cities. Thus, contamination, in particular that of heavy metals, has driven much traditional research on urban soils due to the risks posed to human health (Bullock and Gregory, 1991; Li et al., 2018).

Li et al. (2018) reviewed the range of organic and inorganic pollutants in urban soils and linked these to risks to human health. Studies highlighted by the literature search include those focused on heavy metals (Trammell et al., 2011; McClintock, 2015; Bretzel et al., 2016; Setala et al., 2017); polycyclic aromatic hydrocarbons (PAHs) (Lorenz et al., 2006; Monserie et al., 2009); salts used for road de-icing (Bouraoui et al., 2019); and anthropogenic residues, including traces of actinolite and chrysotile, types of asbestos (Kopel et al., 2016), which further contribute to human health risks.

These studies highlight contaminants present in urban soils, and that reducing public exposure to contamination is crucial. However, they do not typically frame the recycling, degradation and storage of contaminants as an ES provided by urban soil. Thus, while we know the levels at which substances become dangerous to human health, we do not necessarily study the soil's ability to recycle them, store them, and prevent them from being available for human exposure. A small number of studies addressed this, for example, Wang et al. (2015) showed that soil's natural attenuation capacity has strong potential to retain contaminants in urban areas and prevent public exposure; however, attenuation capacity is impacted by urban land use and the extent of soil sealing. More broadly, there is a need to highlight where urban soils are providing this service, protecting humans from exposure, or conversely where the service is compromised, provides a disservice, and urban soils pose a risk.

Carbon storage and GHG regulation

A recent review by Vasenev and Kuzyakov (2018) found that urban soil C content may be higher than in natural soils, and combined with C accumulation through the soil profile to 100 cm, resulted in total C stocks 3-5 times greater in urban soils than natural soils. Across all climates and city sizes, residential areas showed the highest soil organic carbon (SOC) stocks while industrial zones and roadsides showed the highest inorganic C and black C stocks (Vasenev and Kuzyakov, 2018).

Studies identified by the literature search illustrate a comparison between urban and non-urban soils for C storage. Urban park soils in Milan were found to have higher SOC stocks (0-40 cm) compared with croplands in the region, and comparable SOC stocks to other non-urban soils of the region (Canedoli et al., 2020). An analysis of Leicester (UK), including both vegetation and soils, found that urban SOC storage was significantly greater than in surrounding agricultural soils, and that 82% of the city's overall organic C budget was stored in urban soils (Edmondson et al., 2012). However, in Harbin city (China), urban SOC stocks (0-20 cm) were lower than local natural forests (Lv et al., 2016).

Within cities, urban land cover and vegetation type can influence urban soil C. Residential gardens and open spaces were found to have the highest total C stock (0-25 cm) in Madison (USA) by Ziter and Turner (2018) who note the legacy effects of historical land uses on urban soils. In Leicester (UK), residential garden soil had higher SOC concentration than soil in public greenspaces (Edmondson et al., 2014b). Urban soil under trees has been shown to have higher soil C stock (0-30 cm) than soil under grass (Livesley et al., 2016); while Edmondson et al. (2014c) found that SOC enhancement was related to tree species, with SOC being lower under mixed woodland. Urban soil C storage may also be affected by the type of plant litter inputs, for example, greater soil C retention has been suggested as a result of slower decomposition under plants producing recalcitrant litter, such as *Picea abies* and *Calluna vulgaris*, compared to labile litter (Vauramo and Setala, 2011; Setala et al., 2016).

A consistent pattern between urbanisation and soil C has not been found. In Singapore roadsides, SOC was inversely related to urbanisation (Ghosh et al., 2016); while in gardens in Zurich it was found to be positively correlated with urbanisation density (Tresch et al., 2018). A notable impact of urbanisation is soil sealing, and while studies into sealed soil are limited, some illustrate that soil sealing reduces SOC (Wei et al., 2013; Wei et al., 2014a). However, Edmondson et al. (2012) found no difference in SOC storage between greenspace soils and sealed soils at equivalent depths; and Vasenev and Kuzyakov (2018) note that cultural layers and buried horizons can contribute to sealed soil C stores being isolated but not depleted.

Anthropogenic additions and imported fill materials can contribute varying levels of C to soils. For example, Herrmann et al. (2017) found imported fill soils had lower total C content than pre-existing soils, with large variability in the data. Engineering of urban soils has been considered to capture and store soil C using demolition materials. These can be rich in calcium and magnesium which capture atmospheric C through weathering and secondary carbonate mineral precipitation (Washbourne et al., 2012). In addition, black C, arising from incomplete combustion of fossil fuels, can accumulate in urban soil and is considered highly stable, and thus represents an important pool of soil C with long residence times (Canedoli et al., 2020).

Only two of the 125 'quantified' papers measured GHG emissions. The New York City Afforestation Project recorded higher N_2O emissions where shrubs and compost were not incorporated prior to tree planting, highlighting that plant and microbial uptake of inorganic N is important in regulating N_2O losses from urban soils (Pierre et al., 2016). In urban lawns in Melbourne it was found that reducing irrigation and fertiliser helped mitigate GHG emissions in garden systems, however, this needs testing in other soil types and environmental conditions (Livesley et al., 2010).

2.3.2.3 Provisioning services

There was a notable lack of studies on provisioning services, particularly on food production, which is in contrast with most non-urban soil ES literature. As discussed in section 2.3.1, urban food is a developed research area but was not studied specifically as an urban soil ES. This may be related to the common practice of importing materials used for urban food growing, such as compost or topsoil, thus giving the native soil less importance and consideration. Observations of food production were linked more to other services such as wellbeing or biodiversity, rather than solely quantifying the food itself. For example, in a study where radish *Raphanus sativus* were grown, the size of the food growing areas was shown to lack a correlation with the abundance and diversity of invertebrates, suggesting that even small food production sites can still provide ESs related to invertebrates (Biffi et al., 2019). Soil contamination can present a health risk from exposure, either from eating food grown in contaminated soil or from gardening and skin exposure to the soil. Issues around this can be low levels of concern and inconsistent knowledge of gardeners, barriers to conducting soil tests, and limited knowledge of best practice to reduce exposure (Kim et al., 2014).

Only two papers in the literature search considered the physical support of urban soils as an ES, highlighting the hazards of urban soils with poor mechanical properties (Vallone et al., 2008) and risks associated with swelling soils and damage caused to infrastructure, urging the inclusion of soil functionality in urban development (Stell et al., 2019).

2.3.3 Directions for future urban soil ES research

The preceding literature analysis and summary highlight several gaps in knowledge and needs for future research. Here, we summarise a number of research gaps identified and discuss opportunities for future work and collaboration to enhance urban soil ES.

2.3.3.1 Urban soil multifunctionality and trade-offs

Urban areas exhibit high heterogeneity, and potential ES providing areas are required to provide for many and diverse users (Gomez-Baggethun et al., 2013). Enhancing urban soil ES provision is dependent upon the requirements of beneficiaries of those services, as well as the management and treatment of the soil. Nonetheless, given the high density of people living in urban areas and wide range of urban soil ESs it is clear that multifunctionality is key; yet this analysis highlights that within the literature researched (section 2.3.1), only two papers studied four urban soil ESs together (Montgomery et al., 2016; Tresch et al., 2018) and one studied five (Míguez et al., 2020).

To deliver multifunctionality and management win-wins it is necessary to deepen and integrate our understanding of urban soil ESs across disciplines and professions. Opportunities for this could arise though increased study of multiple functions and the inclusion of soil multifunctionality in planning and green infrastructure policy (Scott et al., 2018). These policies could strengthen the protection of existing, and provision of new, urban greenspaces that take account of multiple soil ESs. They could also encourage the protection of existing, and creation of new, urban woodland that aids runoff and stormwater regulation (Ziter and Turner, 2018; Phillips et al., 2019) and soil C storage (Edmondson et al., 2014b; Edmondson et al., 2014c; Setala et al., 2016). Integration of soil ESs into masterplanning and infrastructure projects is necessary to enhance ESs and reduce disturbance to soil functions. Landscape design that enables multiple functions would incorporate diverse vegetation across greenspaces, encouraging a range of microbial and fungal communities and the soil processes they provide (Hui et al., 2017; Tresch et al., 2019b). Management of urban greenspaces also plays a key role, through maintenance schedules and increasing organic matter to enable soil to perform numerous functions (Lorenz and Lal, 2015; Setala et al., 2017). Win-wins may arise through practices such as SuDs to enable water storage, reduce runoff and capture and filter pollutants; or the use of suspended pavement systems and new developments in tree pit design to enable greater water flow whilst also providing bioretention (Tirpak et al., 2019).

However, as urban environments present such complexity, there will be decisions about trade-offs that need to be made. In contaminated soils there may be a choice between the mobility and leaching of contaminants or excess nutrients and improving drainage and infiltration. Choices of vegetation type can influence soil properties and, therefore, urban greenspace planting can influence outcomes for soil functions and service delivery. While these choices have been considered in studies in specific contexts, there remains a gap in clarity over best practice for urban greenspace management and landscape design for the provision of multiple ESs and consideration of trade-offs according to ES requirements for different contexts.

2.3.3.2 Gaps and Opportunities

Beyond multifunctionality, which we identify as an overarching gap, the systematic review allows us to identify six further areas which we believe are key gaps and opportunities for future research.

1. **Water** – whilst much work exists on SuDs and stormwater dynamics, it does not appear to be connected with the ES community. It is vital that urban soil water dynamics are recognised within ES assessments and in considering the benefits of urban green infrastructure. Connections between soil water researchers, SuDs practitioners and the ES community need to be strengthened to enable this important work to be shared. It is also necessary to consider the impacts of soil sealing, compaction and climate change on urban water dynamics for the future.
2. **Food** – interest is increasing in urban agriculture and it is essential that it is connected with the urban soil ES community to ensure the wider benefits of urban soil are known. This will allow consideration of the environmental and social benefits of urban agriculture, as well as risks associated with contamination. It will also enable food growing to be quantified and captured more holistically, and key messages to reach urban planners and policy makers.
3. **Cultural** – in securing these services for the future, it is vital that we capture the importance of urban soils for the range of cultural services it provides, whether through supporting provision of greenspaces for improved mental and physical heath, well-being through food growing, providing aesthetic or spiritual inspiration and sense of place, or through interpreting the layers of history that

soil stores through archaeology. It is especially necessary to take a holistic view of the variety of cultural and wellbeing services that urban soils provide, particularly as urban populations continue to grow, and to take them into account when considering benefits to people.

4. **Global research** – much of the current work is focused on the USA, China or Western European countries. There is a need for research to expand into other global regions, such as Africa, South America and Australia and Oceania, to consider the impacts of urbanisation on soils in a range of climates and in different urban contexts. This is particularly important with increasing pressures on land as cities grow rapidly across the world.
5. **Interconnection between researchers and policy** – there is a need to share quantification methods and findings across research disciplines and communities to enable the vast complexity of ES research to be shared and taken up by practitioners and policy makers. It is important that researchers work together and consider the impact of language and terminology on the uptake of research methods and findings, particularly in relation to planning and policy. This will also aid the study of multiple services and enable the uptake of methods by wider groups, NGOs, businesses and organisations.
6. **Drivers of change** – future drivers such as soil sealing, climate change, and the use of Technosols need to be considered to allow us to gain insight into how urban ecosystems will function as these drivers exert increasing influence. There is also a need to take into account how ESs may be affected by the combined effects of these drivers of change.

2.4 Conclusions

Research into urban soil ESs is a new but growing body of work and is providing much needed information on how urban soils function within the complex, heterogeneous contexts of cities. Most of the research focuses on supporting processes and selected regulating services, such as C storage and recycling of wastes. While the emphasis on supporting processes provides us with data to understand urban soil processes, it does not provide information that is easily used by those outside the soil science community and, thus, taken into urban planning and management. To address this, it is necessary for both supporting processes and ESs to be studied; and research into multifunctionality was highlighted as a key direction for the future. This would also address other gaps found in the literature, such as urban food growing, water dynamics and cultural services rarely being identified as services provided by urban soils. We hope that addressing these gaps will enable urban soils to be better understood and accounted for in the planning, design and management of urban areas in order to support future human wellbeing and urban ecosystem health.

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3. The effects of sealing on urban soil carbon and nutrients

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Abstract

Urban soils are of increasing interest for their potential to provide ecosystem services such as carbon storage and nutrient cycling. Despite this, there is limited knowledge on how soil sealing with impervious surfaces, a common disturbance in urban environments, affects these important ecosystem services. In this paper, we investigate the effect of soil sealing on soil properties, soil carbon and soil nutrient stocks. We undertook a comparative survey of sealed and unsealed greenspace soils across the UK city of Manchester. Our results reveal that the context of urban soil and the anthropogenic artefacts added to soil have a great influence on soil properties and functions. In general, sealing reduced soil carbon and nutrient stocks compared to greenspace soil, however, where there were anthropogenic additions of organic and mineral artefacts this led to increases in soil carbon and nitrate content. Anthropogenic additions led to carbon stocks equivalent to or larger than those in greenspaces; this was likely a result of charcoal additions, leading to carbon stores with long residence times. This suggests that in areas with an industrial past, anthropogenic additions can lead to a legacy carbon store in urban soil and make important contributions to urban soil carbon budgets. These findings shed light on the heterogeneity of urban sealed soil and the influence of anthropogenic artefacts on soil functions. Our research highlights the need to gain further understanding into urban soil processes, in both sealed and unsealed soils, and the influence and legacy of anthropogenic additions on soil functions and important ecosystem services.

3.1 Introduction

To date, little attention has been given to urban soil and its functions, however, the importance of urban soil is increasingly being recognised due to its role supporting sustainable urban development and the provision of soil ecosystem services in cities (Pavao-Zuckerman, 2012; Morel et al., 2015; Yang and Zhang, 2015; Vasenev et al., 2018; Pouyat et al., 2020). In particular, interest has increased in the ability of urban soils to store carbon (C) and contribute to climate regulation, as well as cycling nutrients and supporting urban plant growth (Pouyat et al., 2006; Edmondson et al., 2014a; Herrmann et al., 2017; Setala et al., 2017). The number of people living in urban areas is projected to grow, with almost 70% of the world's population expected to live in urban areas by 2050 (United Nations, 2019). This expansion of urban areas will have consequences for soil and the ecosystem services it's able to provide in urban areas.

Urbanisation leads to highly heterogeneous soils that exhibit a wide range of soil properties (Lehmann and Stahr, 2007) and changes in soil structure such as alterations to soil horizons (Herrmann et al., 2018). It also leads to additions of anthropogenic materials known as artefacts, which include brick, concrete, metals and plastics (Bullock and Gregory, 1991; Lehmann and Stahr, 2007), as well as contamination with heavy metals which can alter nutrient cycles (Zhao et al., 2013), and organic and inorganic pollutants (Li et al., 2018). Increasing urbanisation will lead to increased soil disturbance, contamination and soil sealing with impermeable surfaces, such as roads and pavements (Scalenghe and Marsan, 2009; EU, 2012; Artmann, 2014). Soil sealing commonly involves the removal of topsoil, also known as scalping, where the upper part of the soil is removed down to the subsoil, thus creating a new soil surface (Lehmann, 2006). Sealing also leads to additions of anthropogenic materials to sealed soils, such as road foundation aggregates and other human-made artefacts, before it is then sealed with tarmac, concrete or paving slabs. As a result, sealing is likely to have large effects on soil functioning and the soil-mediated ecosystem services of C storage, water regulation and nutrient cycling.

Recent studies have revealed the notable C storage potential of urban soils (Pouyat et al., 2006; Raciti et al., 2011; Lorenz and Lal, 2015; Vasenev and Kuzyakov, 2018), though less is known on the C storage of sealed soils. Early studies assumed low to zero storage of C beneath sealed surfaces (Bradley et al., 2005). However, more recent research has illustrated that sealed soil C makes an important contribution to wider urban soil C stores (Edmondson et al., 2012; Cambou et al., 2018). Studies indicate that, in general, soil sealing leads to a reduction in soil organic carbon (SOC) (Raciti et al., 2012; Wei et al., 2014a; Wei et al., 2014b; Piotrowska-Długosz and Charzyński, 2015; Vasenev and Kuzyakov, 2018). However, some studies have found similar SOC storage between sealed and non-sealed soils at equivalent depths, indicating that reductions in SOC stocks near the surface were a result of topsoil removal, while stocks in subsoils were no different (Edmondson et al., 2012; Cambou et al., 2018). Inorganic C (IC) also provides an important contribution to C storage in urban soils (Vasenev and Kuzyakov, 2018; Pouyat et al., 2020) however, IC is much less commonly studied in sealed soils. Therefore, the knowledge on soil C storage in sealed soils remains limited and there is a need to further understand the storage potential of sealed soil, the SOC and IC dynamics beneath sealed surfaces and how this contributes to wider urban soil C storage.

Knowledge is also limited on the effects of soil sealing on nutrient stores; how anthropogenic activities may influence these and the subsequent consequences for urban soil nutrient cycling. A number of studies have revealed that sealing generally leads to a reduction in nitrogen (N) storage (Raciti et al., 2012; Wei et al., 2014a; Wei et al., 2014b; Piotrowska-Długosz and Charzyński, 2015; Majidzadeh et al., 2017), and it has been suggested that there is a decoupling of C and N in sealed soils (Raciti et al., 2012; Wei et al., 2014b). The processes involved in nutrient cycling have, however, been less studied. Observations of N mineralisation and nitrification suggest that these processes are reduced by sealing (Zhao et al., 2012; Wei et al., 2014a), while observations of ammonium and nitrate levels have varied (Zhao et al., 2012; Martinová et al., 2016; Majidzadeh et al., 2018). We are yet to have a clear understanding of both the impacts of sealing on N cycling processes or the mechanisms behind these alterations. In addition, there is very limited research on phosphorus (P) in sealed soils, despite this being an important nutrient for plant productivity and water quality. Studies have observed higher Olsen P and extractable P in sealed soils (Wei et al., 2014a; Martinová et al., 2016; Majidzadeh et al., 2017; Majidzadeh et al., 2018); while one study found no difference in available P between sealed and unsealed soil (Piotrowska-Długosz and Charzyński, 2015). Therefore, the effects of urban soil sealing on soil P remain largely unclear. The availability of N and P has implications for SOM mineralisation and C storage, as well as soil nutrient status and leaching, and at present, knowledge on the dynamics between C, N and P in sealed soil is lacking.

There is a need to further our understanding of C and nutrient stocks in sealed soils, to gain a clearer picture of how they contribute to C and nutrient stocks across the wider urban landscape, and what the implications are for urban soil ecosystem services. In this paper, we aim to investigate the effects of soil sealing on urban soils, their properties, and important soil functions including carbon storage and nutrient cycling. We hypothesise that sealed soils will have (i) lower soil C stocks, (ii) lower soil nutrient stocks, and (iii) altered nutrient dynamics compared to greenspace soils. To test these hypotheses, we undertook a comparative survey of soil C and nutrients in sealed and unsealed soils across the UK city of Manchester, constituting one of the largest studies of sealed soil to date. We measured soil pH, bulk density and moisture content to provide insights into soil properties; soil C stocks and extractable organic and inorganic C (hypothesis i); soil N and P stocks (hypothesis ii); and ammonium and nitrate content (hypothesis iii).

3.2 Materials and Methods

3.2.1 Study area

Soils were sampled from Greater Manchester, a metropolitan region in the North West of the UK with a population of 2.8 million (ONS, 2021). The study focused on the wider city area within the M60 motorway and the town of Rochdale within the Greater Manchester Region (Fig. 3.1). The National Soil Map for England and Wales, via the Soilscapes Viewer online (Soilscapes, 2020), shows that the area east of the M60 has slowly permeable, seasonally wet, acid loamy and clayey soils. The south-west has naturally wet and very acid sandy and loamy soils; while the north-west and Rochdale are a combination of slowly permeable, wet acid loamy and clayey soils with areas of floodplain soil with high groundwater, and areas of freely draining slightly acid sandy or loamy soils.

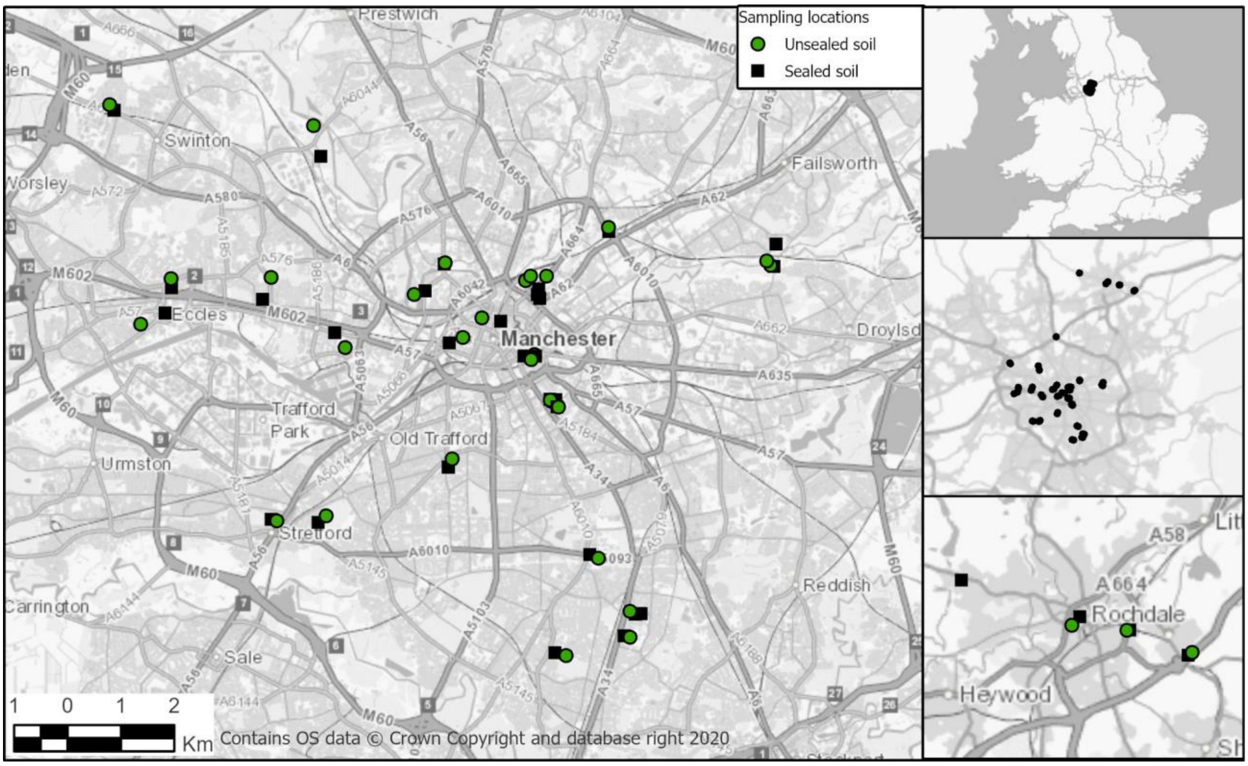

Figure 3.1: Map showing sampling locations across the Greater Manchester Region.

3.2.2 *Soil Sampling*

Sampling was undertaken to allow a comparison of sealed and unsealed greenspace soils. Sampling was undertaken between July and September 2018. Sealed soils were sampled from roadworks or construction sites where work had recently opened up the sealed surface of roads and pavements. As such, the sampling strategy was opportunistic due to the constraints of accessing sealed soils during roadworks activity. Unsealed soils were sampled from the nearest greenspace, park or roadside to the sealed site, and samples were taken from within a grassed lawn area. The distance between sealed and greenspace sites varied with each sample (between 0.25 m – 330 m) and, therefore, they are not considered paired samples. All soils were sampled to a depth of 10 cm of available soil. In greenspaces, soils were sampled from open grassed areas where litter consisted of roots and dead grass leaves. The turf and root mat were removed and the soil was sampled down to 10 cm. In sealed soils, imported construction materials consisted of limestone gravel or chips, construction rubble including brick or concrete, sharp sand, charcoal and ash. Profiles and horizons were not consistent across the sites due to the heterogeneous nature of soil sealing. In general, profiles consisted of a sealed surface, various layers of road or pavement foundation materials, and a clay rich subsoil underneath (see Fig. 10.1, Appendix 3). The depth of construction materials varied from 30-110 cm depth; most samples were collected between 60-80 cm depth, sampling the top 10 cm of available soil under the construction materials. This sampling method allowed a comparison between the top available soil in each profile to understand the properties and functions of each soil. At all sites two samples were collected, one using a metal bulk density core (6 cm diameter), and a second sample using a trowel for additional analyses using fresh soil. Samples were collected in plastic bags, kept in a cool box while transported, and refrigerated until fresh soil analyses were undertaken within one week. A total of 68 sites were sampled, with 36 sealed samples and 32 greenspace samples.

3.2.3 Soil Analysis

3.2.3.1 Urban soil categorisation

Urban soil often contains large amounts anthropogenic additions, or artefacts, which are human-made or derived materials and can include bricks, pottery, glass, crushed stone, charcoal cinders, wood or waste materials. Technosols are defined in the WRB as either containing large amounts of artefacts, having an impermeable geomembrane or having a technic hard material at the soil surface, such as concrete, asphalt or worked stone (FAO, 2015). All the sealed soil samples collected are considered Technosols as a result of the continuous hard sealed surface. However, we observed that some sealed samples contained numerous artefacts while others did not and, apart from being sealed, appeared relatively undisturbed by human activity. These artefacts appeared to be fragments of the materials used in road or pavement construction (see section 3.2.2) which had disintegrated and been mixed into the soil. The addition of artefacts can considerably alter the properties and functions of soil, such as water holding, C storage or nutrient status. Therefore, the sealed samples were categorised into two types: those relatively undisturbed other than by sealing, herein referred to as sealed undisturbed soils (SU); and those with notable additions of anthropogenic artefacts, herein referred to as sealed anthropogenic soil (SA) (Fig. 3.2).

Wet sieving was undertaken on subsamples of the sealed soils to distinguish between SU and SA soils. We used the proportion of material in the >200 μm fraction to determine the level of anthropogenic additions and serve as a proxy for the proportion of artefacts. Soils with visible artefacts exhibited more than 40 % of subsample mass in the >200 μm fraction; thus, subsamples with more than 40 % mass in the >200 μm fraction were classed as SA soils, and those with less than 40 % in the >200 μm fraction were classed as SU soils. The fragmentation of artefacts into smaller fractions made it impractical and inaccurate to use a measure the mass of artefacts alone. Using material >200 μm served to describe the samples well and enabled a consistent comparison between anthropogenic and undisturbed soils.

Figure 3.2: Diagram showing soil profiles and sampling depths of the three soil categories.

3.2.3.2 *Soil preparation and properties*

As urban soil commonly contains imported anthropogenic materials the in-situ volume, density and structural properties of the soil are related to these materials, and in some cases, they make up a large proportion of the soil. As such, we deemed it appropriate not to remove the coarse fraction above 2 mm prior to bulk density calculation to provide a representation of the in-situ soil. It is important to note, however, that using these bulk density measurements with coarse fractions may lead to an overestimation of carbon stock values if the soil contains many gravel sized artefacts. Bulk density cores were weighed, dried at 105 °C for 48 hours (due to high clay content) then re-weighed to enable calculations of bulk density and gravimetric soil moisture. They were then used to measure the proportion of material >200 μm by sieving to determine soils with anthropogenic additions.

The second set of samples were used for the fresh soil analyses of pH, nitrate and ammonium extraction. For pH analysis, 10 g fresh soil was mixed with 25 ml distilled water, shaken on an orbital shaker for 30 minutes at 180 rpm and left to settle for 30 minutes. The pH was then measured with a pH probe (Mettler Toledo, SevenCompact S220) at the soil-water interface. The remaining fresh sample was dried at 70 °C for 48 hours. It was then homogenised using a pestle and mortar, passed through a 2 mm sieve, and subsamples were taken for analyses of total P, extractable OC and inorganic C. Further subsamples were then dried to 105 °C prior to CN analysis and loss on ignition. Many samples of sealed soil were high in clay and for this reason the method deviates from the traditional approach to sieving the samples prior to drying.

3.2.3.3 *Carbon and Nutrient Analysis*

To determine total C and N concentrations samples were dried at 105 °C for 24 hours, ball-milled to a powder and analysed for total C and total N concentration and CN ratio using a dry combustion CN analyser (El Vario analyser, Elementar, Hanau, Germany). Soil organic matter (SOM) was estimated using the loss on ignition (LOI) method described by Heiri et al. (2001). Samples were dried at 105 °C for 24 hours prior to weighing, heating at 550 °C for 6 hours and re-weighing to determine the loss of SOM by weight as a percentage.

Organic C (OC) and inorganic C rendered extractable to 0.5M K_2SO_4 were measured, as described by Vance et al. (1987) without fumigation with $CHCl_3$ so as to measure non-microbial biomass C. Briefly, the acidity of the K_2SO_4 was checked and adjusted to between pH 6.8–7 using NaOH. For the extraction, 5 g of dry soil was mixed with 20 ml of K_2SO_4 (0.5M) and shaken on an orbital shaker for 1 hour at 180 rpm. It was left to settle for 10 minutes then filtered through a Whatman no. 42 filter. Filtrate was diluted 1 part to 8 parts MQ water and analysed for extractable total and inorganic C in a TOC analyser (Shimadzu TOC-L_{CPN} TN). Extractable OC is determined as total extractable C minus extractable inorganic C.

Ammonium and nitrate pools were measured with the aim of understanding mineral N content and dynamics. The extraction was undertaken using 1M KCl as an extraction matrix (Kachurina et al., 2000; Saha et al., 2018). For extractions, 5 g of fresh soil was mixed with 25 ml KCl (1M) and shaken on an orbital shaker for 1 hour at 180 rpm. It was filtered through a Whatman no. 6 filter and filtrate was analysed for nitrate and ammonium using a colorimetric segmented flow analyser (AA3, Seal Analytical, Southampton, UK). Total phosphorus (P) content was analysed using a sulphuric acid / hydrogen peroxide digestion method by Rowland and Grimshaw (1985). For the digest, 0.2 g of dry ground soil was mixed with 4.4 ml digest reagent and heated gently until the vigorous reaction had subsided. Heat was increased to 400 °C and boiled for 2 hours until the digest had cleared. Once cool samples were diluted to 50 ml with MQ water and filtered using a Whatman no. 6 filter. Filtrate was diluted a further five times and analysed for total P on a colorimetric segmented flow analyser (AA3, Seal Analytical, Southampton, UK). Soil total C, N and P stocks were calculated using bulk density values to a depth of 10 cm and C, N and P contents, as per the guidance from the FAO (2018).

3.2.4 Statistical Analysis

Analysis was undertaken to determine the difference between sealed and greenspace soils, and also to determine whether there was a difference between the two categories of sealed soil, SU and SA soils. Data for the majority of variables did not exhibit a normal distribution according to the Shapiro-Wilks test. Therefore, all datasets were analysed using the non-parametric Kruskal Wallace test to identify significant differences between the three categories of soil, and the Dunn-Bonferroni post hoc test to determine where the significant difference was between soil categories. All analyses were undertaken in SPSS 26, and figures were produced using R version 4.1.0.

3.3 Results

Sealed samples were categorised into two groups, those that were relatively undisturbed other than by sealing, referred to as sealed undisturbed (SU) soils, and those with notable additions of anthropogenic material, referred to as sealed anthropogenic (SA) soils. Of the 36 sealed samples, 22 were classed as SU soils and 14 were classed as SA soils. The results of the analyses are summarised in Table 9.1 in Appendix 2. Results are presented for soil properties, soil C and SOM analyses, nutrient contents and stoichiometry respectively.

3.3.1 Soil Properties

Figure 3.3 summarises the soil properties across the sealed and greenspace categories. Both categories of sealed soil had significantly higher pH than the greenspace soil (p <0.001); while both had lower soil moisture, though only SA soil was significantly lower than greenspace soil (p = 0.006). Both pH and soil moisture data showed no significant difference between the two sealed soils, however, all three soils exhibited significantly different bulk densities (p <0.001) with SU soils having the highest bulk density and SA soil the next highest (Fig. 3.3 a, b).

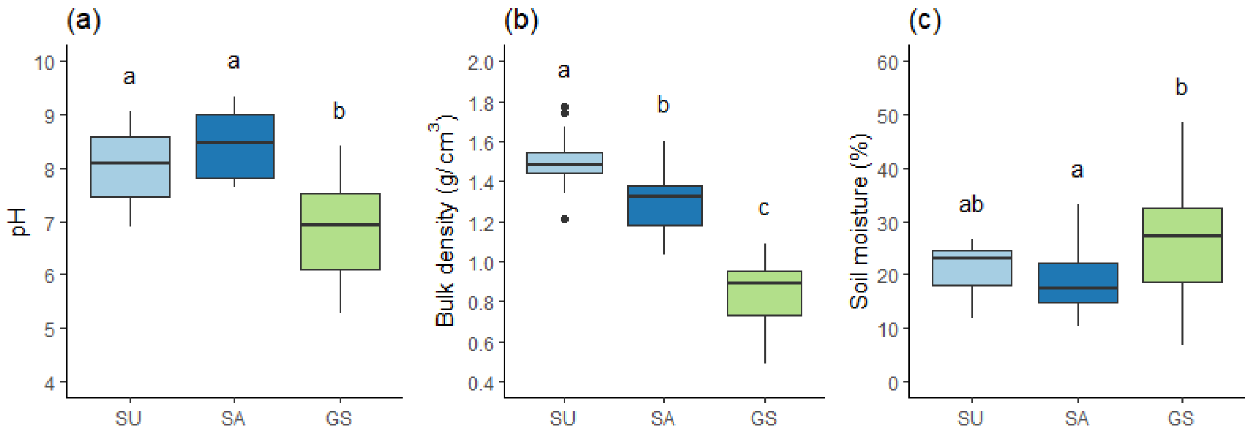

Figure 3.3: Soil properties: (a) pH, (b) bulk density (g/cm^3) and (c) soil moisture content (%) for sealed undisturbed (SU; n=22), sealed anthropogenic (SA; n=14), and greenspace (GS; n=32) soils. Boxplots show upper and lower quartiles, whiskers show upper and lower most values, and horizontal line shows the median. Circled data points show outliers at 1.5 times the IQR. Different letters indicate a statistically significant difference between soil categories at the 0.05 level.

3.3.2 Carbon

Total C data indicated that SU soil had significantly lower total C concentration than both other soils (*p* <0.001), while SA soil was statistically no different to greenspace soil. Total C stock for the top 10 cm of soil exhibited a significant difference across all three soil categories (*p* <0.001) (Fig. 3.4 b), with SA soil having significantly greater total C stock than greenspace soil (8.06 kg m^{-2} ± 4.65 and 4.92 kg m^{-2} ± 1.11 respectively), and greenspace soil having significantly greater stock than SU soil (3.10 kg m^{-2} ± 1.45). SU soil had 37 % less total C stock than greenspace soil, while SA soil showed an increase of 64 % total C stock on that of greenspace soil.

By contrast, SOM data indicated that both sealed soils had significantly lower SOM concentration than greenspace soil (*p* <0.001), and while SA did have greater SOM concentration than SU soil (7.06 % ± 3.20 compared to 5.00 % ± 1.29), there was no statistically significant difference between them (Fig. 3.4 c). SU soil had 55 % less, and SA soil 37 % less, SOM than greenspace soil. SA soils exhibited more variability in Total C stock data than in SOM values, with S.D. 4.65 for total C stock, and S.D. 3.20 for SOM concentration.

Extractable C (K_2SO_4) was analysed to investigate the amount of extractable organic and inorganic C in the samples. In the sealed soils, 76-79 % of the extractable C was organic C, and 21-24 % was inorganic C. In the greenspace soils, 98 % of extractable C was organic C while only 2 % was inorganic C (Fig. 3.4 f). Both sealed soils exhibited significantly lower extractable organic C than greenspace soils (*p* <0.001), with no significant difference between the two sealed soils (Fig. 3.4 d). Extractable inorganic C was significantly higher in both sealed soils compared to greenspace soil (*p* <0.001), with, again, no significant difference between the two sealed soils.

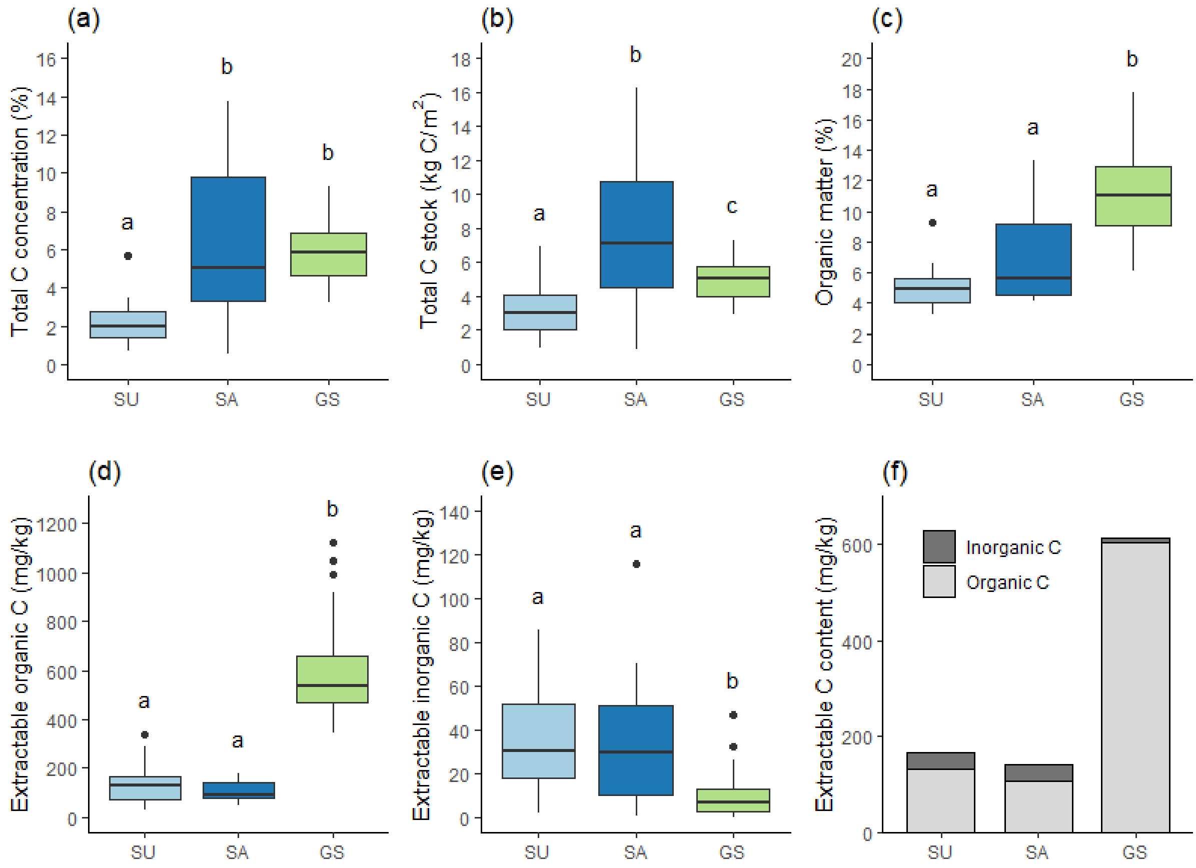

Figure 3.4: Carbon measurements: (a) total C concentration; (b) total C stock; (c) organic matter concentration; (d) K_2SO_4 extractable organic C content; (e) K_2SO_4 extractable inorganic C content; and (f) mean K_2SO_4 extractable total C content; for sealed undisturbed (SU; n=22), sealed anthropogenic (SA; n=14), and greenspace (GS; n=32) soils. Stocks are calculated for the top 10 cm of available soil. Different letters indicate a statistically significant difference between soil categories at the 0.05 level.

3.3.3 Nutrients

Both sealed soils exhibited significantly lower total N stock than greenspace soils (p <0.001), while the two sealed soils showed no significant difference between them (Fig. 3.5 a, b). SU soil had the lowest total N stock (81.20 g m^{-2} ± 22.47) followed by SA soil (90.11 g m^{-2} ± 28.38), with greenspace soil having the greatest total N stock (115.44 g m^{-2} ± 26.49). Total P stock showed a similar pattern, with both sealed soils exhibiting significantly lower total P stock than greenspace soil (p = 0.003) (Fig. 3.5 e, f). SU soil had the lowest total P stock (39.62 g m^{-2} ± 20.91), followed by SA soil (39.42 g m^{-2} ± 23.68), and then greenspace soil with the greatest total P stock (62.62 g m^{-2} ± 36.67).

Analysis of ammonium and nitrate pools (NH_4^+ and NO_3^-) illustrated that SU soil contained greater ammonium content than SA or greenspace soils (Fig. 3.5 c), with a mean of 11.06 mg kg^{-1} (±15.52) compared to 2.53 mg kg^{-1} (±4.88) and 6.10 mg kg^{-1} (±11.74) respectively; however, there was no statistically significant difference in ammonium between the three soils (p = 0.100). Conversely, SU soil exhibited significantly lower nitrate content compared to greenspace soil (p <0.001), while there was no significant difference in nitrate between the SA and greenspace soil (Fig. 3.5 d).

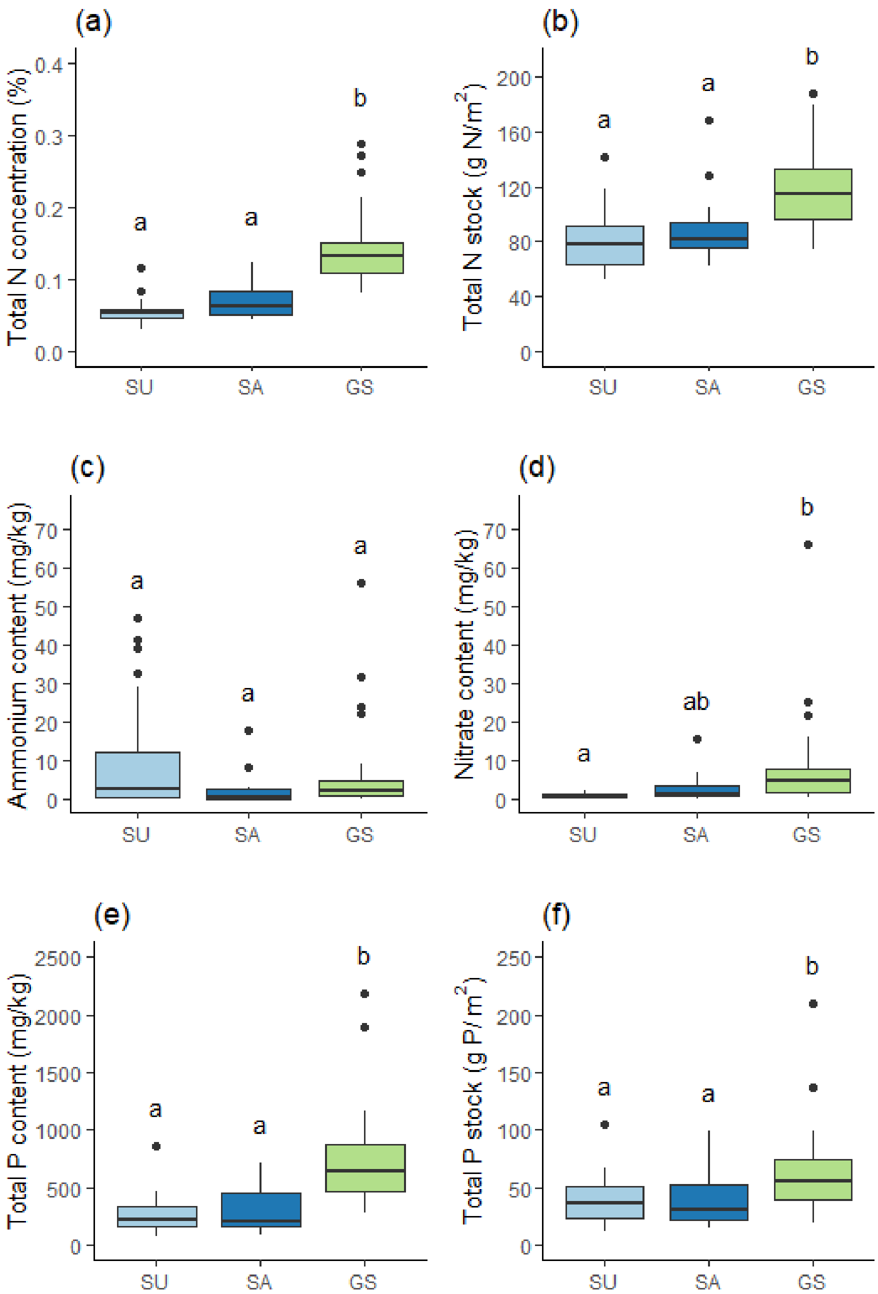

Figure 3.5: Soil nutrients: (a) total N concentration; (b) total N stock; (c) ammonium content; (d) nitrate content; (e) total P content; and (f) total P stock; for sealed undisturbed (SU; n=22), sealed anthropogenic (SA; n=14), and greenspace (GS; n=32) soils. Stocks are calculated for the top 10 cm of available soil. Different letters indicate a statistically significant difference between soil categories at the 0.05 level.

3.3.4 *Stoichiometry*

The C:N ratio was significantly higher in SA soil than SU or greenspace soil (p <0.001), and C:N ratio data for SA soil exhibited much greater variability than that of SU or greenspace soil (SD = 40.45; 12.44 and 6.99 respectively). There was no significant difference between the C:N ratios of SU and greenspace soils (Fig. 3.6 a). The C:P ratio showed a similar pattern, with SA soil having a much higher C:P ratio than SU or greenspace soil (p <0.001); while there was no significant difference between the SU and greenspace soils (Fig. 3.6 b). There was no significant difference in N:P ratio between the three soil categories (Fig. 3.6 c).

Correlation analysis highlighted how SA soil differed from SU and greenspace soil in the relationship between total C and total N, and total C and total P. In SA soil, the C:N and C:P ratios were both significantly greater than those in SU and greenspace soil (Fig. 3.6 a, b); while the N:P ratio showed no significant difference across all three soils (Fig. 3.6 c). There were significant positive correlations between total C and N for all three soils (p <0.001). Figure 3.6 d shows the difference in correlations, illustrating that in SA soil, total C increased markedly though total N did not; while in greenspace soil, both total C and N increased in concordance. SU soil exhibited a positive relationship between total C and N to a lesser degree. A similar pattern was seen in total C and total P correlation (Fig. 3.6 e), however SU soil exhibited a similarly positive relationship between total C and P to that seen in greenspace soil (p <0.001 and p = 0.019 respectively). SA soil did not show a significant correlation between total C and P. Correlation between total N and total P (Fig. 3.6 f) illustrated relatively strong positive correlations in both SU and SA soils (p <0.001 and p = 0.007 respectively), though no significant correlation was seen in greenspace soil.

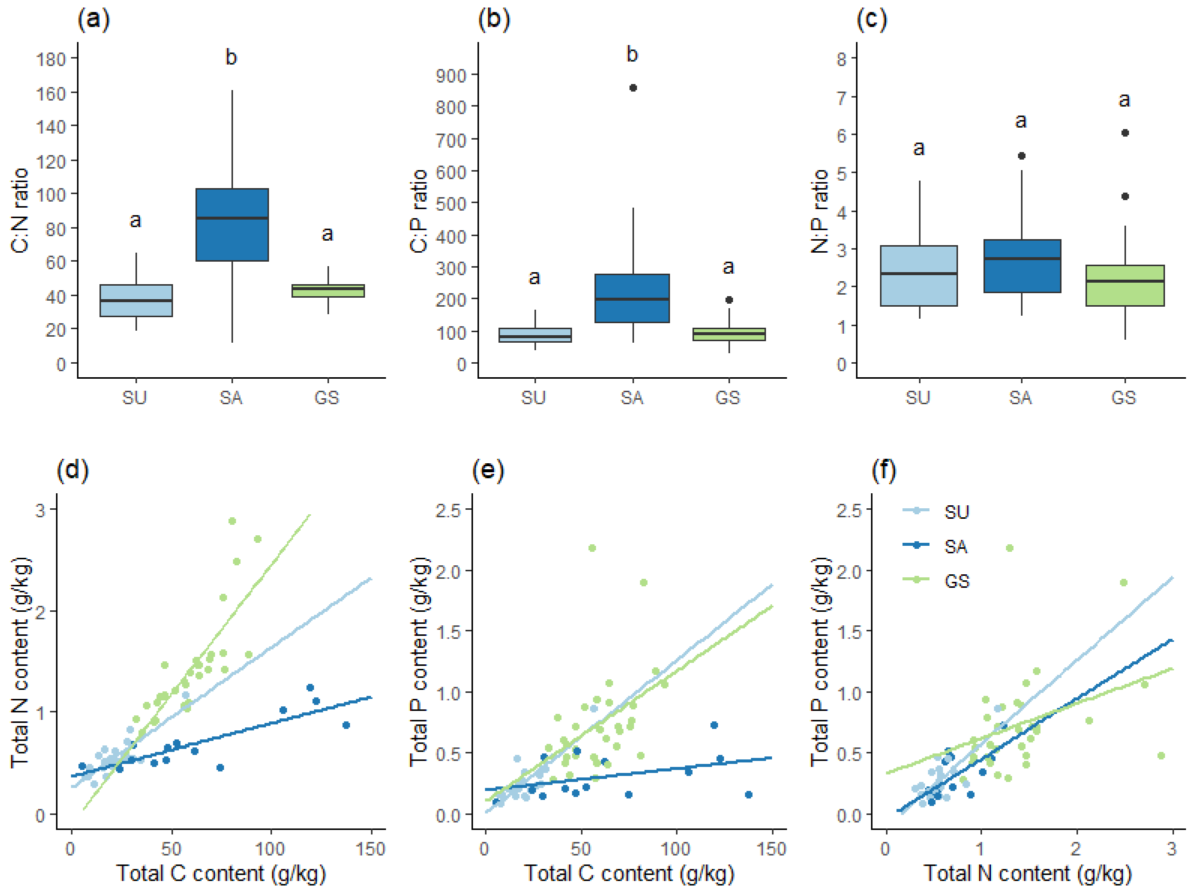

Figure 3.6: Stoichiometry: (a) C:N ratio; (b) C:P ratio; (c) N:P ratio; correlations of (d) C and N, (e) C and P, and (f) N and P; for sealed undisturbed (SU; n=22), sealed anthropogenic (SA; n=14), and greenspace (GS; n=32) soils. Different letters indicate a statistically significant difference between soil categories at the 0.05 level.

3.4 Discussion

3.4.1 Sealed soil C stocks

We set out to test hypothesis i, that sealed soils will have lower C stocks than greenspace soils. The sealed soils (SU) exhibited significantly lower C stocks than greenspace soils (section 3.4.1.2). However, in sealed soils with anthropogenic additions (SA), C stocks were greater than in greenspace soils (section 3.4.1.1). Therefore, the hypothesis was rejected given that not all sealed soils exhibited lower soil C stocks. However, the context and treatment of the soil was important as sealed soils without anthropogenic additions had significantly lower C stocks.

3.4.1.1 SA soil and anthropogenic additions

We found that where there are additions of anthropogenic material to sealed soils, they can provide notably large soil C stores. Our results indicate that SA soil had much greater total C stocks than greenspace soil, and also greater SOM contents than SU soil, though this was not statistically significantly. SA soil had total C stock of 8.06 kg C m^{-2} between 0 and 10 cm depth which is markedly greater than published data for sealed soils to date, while SU soil stored 3.10 kg C m^{-2}, which is comparable, while still larger, than most existing sealed soil C observations. Previous studies have observed stocks of 2.35 kg OC m^{-2} between 0-20 cm in Nanjing City, China (Wei et al., 2014a); 2.29 kg OC m^{-2} between 0-15 cm in New York (Raciti et al., 2012); and 1.25 kg C m^{-2} between 0-10 cm in Alabama, USA (Majidzadeh et al., 2017). In Leicester, UK, Edmondson et al. (2012) found stores of 6.7 kg OC m^{-2} between 40-100 cm under roads, which at equivalent depths to this study would also represent smaller C stocks than SA soil. It is likely that the large discrepancy between SA stocks and previous observations is due to the anthropogenic additions of C to the SA soil. This suggests that materials used in road construction and artefacts added to the soil can contribute significant amounts of C to sealed soil. This flow of materials into soil is commonly cited as a key characteristic of urban soils, in particular Technosols, and leads to the mixing of these materials into soil horizons (Bullock and Gregory, 1991; Weil and Brady, 2017; Herrmann et al., 2018). This highlights the need to consider history, land use and archaeology alongside soil science when studying urban soils (Lehmann and Stahr, 2007; Ziter and Turner, 2018). The influence of human activity on urban soil has been termed a 'cultural layer' which can contribute to urban soil C stores (Vasenev et al., 2013), and, as illustrated by our results, can create a legacy of C storage.

In contrast to the high total C values, SA soil had lower SOM and extractable OC content than greenspace soil. This suggests that some of the total C measured was not detected in the SOM or extractable OC analyses and, thus, was a more stable form of OC or inorganic C. During the sealed soil categorisation (section 3.2.3.1) anthropogenic artefacts were found which would contribute to stable OC or inorganic C in the soil, such as charcoal, concrete and limestone rubble. Charcoal cinders were historically a plentiful waste product from industry and coal power stations in the UK through the nineteenth and twentieth centuries, and were commonly used as a base layer on top of subsoil in road construction in the UK and US (MacBride, 2013). This stable OC would not have been detected in extractable OC analysis; and it may also not have been completely combusted at the temperatures used in LOI as recalcitrant OC, such as black C, can burn to approximately 600 °C (Edmondson et al., 2015). These charcoal cinder additions provide similar recalcitrant OC storage to that of black C, which is ubiquitous in unsealed urban soils as a result of traffic and fossil fuel burning (Hamilton and Hartnett, 2013). Black C plays an important role in soil C storage due to its long residence time and its protection from rapid decomposition (Kuzyakov et al., 2014; Lehmann et al., 2015), allowing it to contribute to stable long-term urban C stores. The contribution of black C to urban soil is a small but important area of study (Rawlins et al., 2008; Edmondson et al., 2015; Schifman et al., 2018), however, at present little is known about black C or charcoal within sealed soils. Our results illustrate that historical human activity may have contributed notable amounts of stable OC to sealed soils in the UK and possibly the US, and that these likely make a significant contribution to urban soil stable C stores in these areas. This has also been observed in New York, where Technosols formed from coal ash exhibited much higher OC stocks than other soils, illustrating that human transported materials can be a source of high OC stocks (Cambou et al., 2018). However, further work on the stability of sealed soil OC is needed to fully understand the C dynamics and long-term impacts on C storage.

Some of this additional stable C may also have come from recalcitrant IC sources. IC, such as carbonate, does not thermally decompose until reaching temperatures of approximately 700-800 °C (Washbourne et al., 2012; Edmondson et al., 2015) and thus would also not be captured in LOI analysis. In addition, K_2SO_4 extraction of IC may not have extracted all recalcitrant IC in the sample. We found high IC content in our sealed samples, consistent with other observations of urban soils, which is due to the weathering of calcium minerals from concrete (Washbourne et al., 2015; Weil and

Brady, 2017) and the use of calcareous materials such as cement-based rubble and limestone in road subbase layers, which add calcium and carbonates to urban soil (Shaw and Reeve, 2008; Kida and Kawahigashi, 2015; Asabere et al., 2018). Soil IC stocks make an important contribution to sealed soils and subsoils in cities, highlighting their importance as hidden stocks in C assessments (Vasenev and Kuzyakov, 2018). The importance of calcium rich minerals and dissolved carbonates in urban soil have been highlighted for the removal of CO_2 to form calcium carbonate (Washbourne et al., 2015; Jorat et al., 2020); a process that is also being promoted for agricultural soils as a carbon capture mechanism (Beerling et al., 2020). While little is known about water or air flow under sealed surfaces, this process has been observed in sealed soil where cracks in paving have allowed water to infiltrate, and dissolved calcium reacted to form calcium carbonate which then moved into deeper soil horizons (Kida and Kawahigashi, 2015). At present, the extent to which this process is occurring in sealed soils is unknown.

3.4.1.2 SU soil compared to greenspace soil

The SU soil had been sealed and remained largely undisturbed and altered by human activity following sealing. In comparison with greenspace soil, SU soil exhibited reduced C stocks, a pattern that has been seen in most other studies of sealed soil (Raciti et al., 2012; Wei et al., 2014a; Wei et al., 2014b; Majidzadeh et al., 2017; Majidzadeh et al., 2018). Our results showed a reduction of 37 % of C stock in SU soil compared to greenspace soil, a notably smaller reduction than that seen in other studies, where reductions were 66 % in New York (Raciti et al., 2012); 68 % in China (Wei et al., 2014b); and 61.86 % (Majidzadeh et al., 2017). However, a review by Vasenev and Kuzyakov (2018) found that on average, at 50-100 cm depth, sealed soil OC stocks were 25 % lower than under lawns, but 10 % higher than under trees and shrubs. This suggests that C losses may be smaller further down the soil profile, and additionally, that the context of the unsealed greenspace soil is important when making comparisons. Indeed, many greenspace soils are also influenced by urban land use and anthropogenic additions, whether directly or indirectly, and may also exhibit altered C stocks as a result (Ziter and Turner, 2018; Canedoli et al., 2020). Despite our results showing smaller soil C stocks in SU soil than greenspace soil (3.10 kg C m^{-2} and 4.92 kg C m^{-2} at 0-10 cm), our SU soil C stocks were still greater than other published sealed C stocks (reported in section 3.4.1.1); and were comparable to those reported for greenspaces in Alabama, at 3.38 kg C m^{-2} at 0-10 cm (Majidzadeh et al., 2017), and Nanjing City, at 4.52 kg C m^{-2} at 0-20 cm (Wei et al., 2014a).

Due to the excavation of topsoil for road construction, the sealed soil studied is typically from deep in the soil profile. Deep urban soils beyond 50 cm depth are rarely studied, particularly so for sealed soils, however they form a very large proportion of the urban soil profile and play an important role in urban soil C storage (Cambou et al., 2018; Vasenev and Kuzyakov, 2018). Our larger sealed soil C stocks may be a result of various factors. It has been suggested that sealing prevents decomposition due to sealed soil being isolated from the atmosphere and creating unfavourable conditions for microbes (Raciti et al., 2012; Piotrowska-Długosz and Charzyński, 2015) and thus, while sealing may isolate subsoil C stocks from litter inputs and decomposition, it does not necessarily deplete them (Vasenev and Kuzyakov, 2018). It is likely that the SU soil C stocks are a result of the high clay content of the sealed soil, as clay soils can provide high C stabilisation due to organo-mineral complexes which protect C from decomposition (Hassink, 1997; Six et al., 2002; Lorenz et al., 2008). However, the addition of anthropogenic C to road subbases, as previously discussed for SA soil, may also have contributed to the SU soil C stocks through transport of C. Little is known about the movement of water and dissolved nutrients through sealed soil, though it has been suggested that dissolved C may travel into sealed soils from more C rich unsealed areas (Majidzadeh et al., 2017; Pereira et al., 2021); and dissolved charcoal is known to be mobilised in soils where there is water flow (Jaffé et al., 2013). Overall, the findings for SU soil C stocks support the argument that sealed subsoil C plays an important role in urban soil C storage and should be included in urban soil C assessments.

3.4.2 Nutrient stocks are lower in sealed soils

Stocks of total N and total P were significantly smaller in both sealed soils compared to greenspace soil. Therefore, we can accept hypothesis ii, that sealed soils have lower soil nutrient stocks than greenspace soils. The findings for N stocks corroborate those of other sealed studies, where N content and stocks have been consistency lower than in unsealed soils (Raciti et al., 2012; Wei et al., 2014a; Wei et al., 2014b; Piotrowska-Długosz and Charzyński, 2015; Majidzadeh et al., 2017). This is potentially due to the loss of N as a result of topsoil removal during the sealing construction process and the consequent lack of litter inputs following sealing. The reduction in plant growth and organic matter inputs will lead to low levels of substrate and low rates of mineralisation and nitrification, impacting nutrient stocks. It has also been suggested that N content may be reduced in sealed soils due to aqueous losses of dissolved N, or gaseous losses as a result of denitrification (Raciti et al., 2012).

The C:N ratio was notably higher in SA soil than both other soils, likely as a result of additions of anthropogenic C, and correlation analysis showed there were significant positive relationships between C and N in both sealed soils and greenspace soil. This is in contrast to previous studies that found no relationship between C and N in sealed soil (Raciti et al., 2012; Majidzadeh et al., 2017); while one study found sealing led to a lower C:N ratio, with lower C explained by a lack of organic matter inputs, and a disruption in the relationship between C and N (Wei et al., 2014b). It has been suggested that sealing decouples C and N (Raciti et al., 2012; Wei et al., 2014b), an assertion which is supported by the C:N ratio seen here in SA soil, however, the strong positive correlation between C and N in both SU and SA soil suggest this decoupling may not always occur in sealed soils.

Studies of total P stocks are rare in sealed soils. We found that sealing significantly reduced total P stocks in both sealed soils compared to greenspace soil. These findings are contrary to other observations of P, where Olsen P concentration was greater in sealed soils (Wei et al., 2014a; Martinová et al., 2016); and P extracted using the

Mehlich-1 method was greater in soils of crawl spaces beneath houses than in adjacent lawns (Majidzadeh et al., 2017; Majidzadeh et al., 2018). These observed increases have been explained by the absence of P uptake by plants, and reduced loss of P by leaching or runoff (Wei et al., 2014a; Majidzadeh et al., 2018). In addition, P may be higher in some studies where sealing has occurred more recently, as urban greenspace soils can have high P contents (Qin et al., 2019), however, the length of time sealed was not included in this study. In a study in Poland, no difference in available P content was observed between sealed and unsealed soil, though semi-pervious soil did have a slightly lower P content (Piotrowska-Długosz and Charzyński, 2015). Our results may differ from other studies due to the difference in analysis method and the form of P analysed. In addition, some of the differences may be attributable to notable climatic differences between the study locations.

3.4.3 Sealed soil nutrient dynamics

We set out to test hypothesis iii, that sealed soils will exhibit altered nutrient dynamics compared to greenspace soils. In sealed undisturbed soils (SU) mineral N dynamics were altered, with significantly lower nitrate content and higher ammonium content, though this was not significant. However, in SA soils with additions, the nitrate and ammonium contents were comparable to greenspace soils and the effect of sealing appeared to be mediated. Therefore, hypothesis iii was rejected given that nutrient dynamics did not appear to be altered in all sealed soils. However, the context of the soil was important as nutrient dynamics did appear to be altered in SU soils.

The presence of ammonium in SU soil suggests that mineralisation had occurred in this soil to some degree, however, it is unknown whether this had occurred previously, utilising SOM leftover prior to sealing, or whether the process was ongoing. Potential N mineralisation has been observed to be significantly lower in sealed soil (Zhao et al., 2012), though some have found no significant difference in N mineralisation or inorganic N levels between sealed and unsealed soil (Wei et al., 2014a). It has been suggested that nutrient deficiency in sealed soils may stimulate microbes to decompose any available N into ammonium for their survival, and that anaerobic conditions may promote the conversion of nitrate to ammonium by nitrate reductase or to N_2O gas through denitrification (Norton and Stark, 2011; Raciti et al., 2012; Zhao et al., 2012). Ammonium can also accumulate in soil as a result of sorption to clay minerals (Sahrawat, 2008; Nieder et al., 2011; Weil and Brady, 2017); and exchangeable ammonium has been seen to increase following vegetation removal, perhaps due to reduced plant uptake, death of root material and decreased transpiration leading to more water movement in soil (Page, 2004), processes which may also occur in sealed soil.

Average nitrate content for greenspace and SA soils were similar to previous urban soil nitrate observations, which range from 3.3 mg kg^{-1} for bare soil in Beijing (Zhao et al., 2012), to 8.7 mg kg^{-1} for park soil in Leuven, Belgium (Martinová et al., 2016). There was variation between the two sealed soils, with SU soil having significantly lower nitrate content than greenspace soil and SA soil showing no significant difference to greenspace soil. In the SU soil, it is possible that sealing conditions restricted the growth of the microbial community (Lorenz and Lal, 2009), thus preventing nitrification. SU soils exhibited high bulk density suggesting the soil is compacted and may be limited in oxygen. The presence of oxygen is a key control in nitrate production (Sahrawat, 2008; Weil and Brady, 2017), and compacted soils typically have lower aeration, soil moisture and reduced rates of nitrification (De Neve and Hofman, 2000). Our data for SU soil supports findings from previous sealing studies, where nitrification and other microbial activities were notably reduced by sealing (Zhao et al., 2012; Wei et al., 2014a; Pereira et al., 2021). There is also the possibility that our low nitrate content was a result of losses due to denitrification or leaching of dissolved mineral N, as suggested by Raciti et al. (2012). The high pH of the SU soil and the presence of redoximorphic features exhibited by these samples both suggest reduction conditions and anoxic patches, within which denitrification could occur. As such, the low nitrate content in SU soil may have been due to reduced nitrification or increased denitrification, both of which can occur at low oxygen concentrations (Norton and Stark, 2011).

Conversely, the SA soil exhibited slightly higher nitrate content than SU soil and showed no difference to greenspace soil. This may be a result of numerous factors. Bulk density was lower in SA soil suggesting improved aeration and greater oxygen levels than in the SU soil. This could lead to conditions sufficient for nitrification in SA soil leading to the slightly higher nitrate levels, or alternatively, the reduction of denitrification conditions which would lead to reduced nitrate losses as N_2O gas. In addition, charcoal added to soil has been found to alleviate factors that inhibit nitrification (Abdelrahman et al., 2018), suggesting there may be benefits for nitrate levels from the anthropogenic additions of charcoal to the SA soil. As seen in our findings, soil nutrient dynamics within sealed soils remain largely unknown, highlighting the importance of further research into sealed soil processes and the potential effects of anthropogenic materials on these important soil functions.

3.5 Conclusions

This study has found a number of widespread effects of sealing on soil properties, carbon storage and nutrient dynamics. We set out to test the hypotheses that sealed soils would have (i) lower soil C stocks, (ii) lower soil nutrient stocks, and (iii) altered nutrient dynamics compared to greenspace soils. Soil properties were significantly affected by soil sealing, leading to higher pH, lower soil moisture and higher bulk density than in greenspace soils. Sealed undisturbed soils had lower carbon stocks than greenspace soils. However, this study has highlighted for the first time, the potential importance of anthropogenic additions to sealed soil carbon stocks. We found that anthropogenic additions of C-rich materials to sealed soils can lead to notably large soil C stocks, in some cases larger than greenspace soils, forming a legacy C store under sealed surfaces. As such, hypothesis i was rejected given that not all sealed soils exhibited lower soil C stocks. This highlights that land development history is potentially an important control on urban soil C storage and its heterogeneity within, and between, cities. Our findings also indicate that this legacy C storage in sealed soils may include stable OC with long residence times as a result of historic OC additions in areas with an industrial past. However, further work into anthropogenic additions and the long-term OC stability and storage capabilities of sealed soil is needed to provide more detailed information. Further research into the effect of anthropogenic additions on OC stability and storage across the wider urban landscape, including greenspace and sealed soils, would provide a more complete picture of urban soil OC storage. Inorganic C also contributes to the legacy C store in sealed soils due to the weathering of minerals from concrete and calcareous materials; and the potential for atmospheric CO_2 removal due to calcium carbonate production in sealed soils is another area in need of further investigation. Sealed undisturbed soils had altered mineral N dynamics, exhibiting lower nitrate and higher ammonium content than greenspace soils. However, sealed soils with anthropogenic additions had similar nitrate and ammonium contents to greenspace soils. As such, hypothesis iii was rejected given that nutrient dynamics were not altered in all sealed soils, and the additions appeared to mediate the effect of sealing. This may have been a result of improved soil structure and conditions for nitrification, or a lack of conditions leading to denitrification compared to relatively undisturbed sealed soils. Where sealed soils remained relatively undisturbed and altered by human additions, carbon, N and P stocks were all lower, and as such, hypothesis ii was accepted. Overall, this study points to a need to understand how land development history influences sealed soil functioning, and for further studies that advance our understanding of carbon stocks, carbon stability and nutrient dynamics in sealed soils of different contexts.

3.6 Acknowledgements

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4. The persistence of soil carbon across urban greenspaces and sealed surfaces

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Abstract

Urban soils are of increasing interest for their ability to store large amounts of organic carbon. In urban areas, soils are affected by urbanisation in numerous ways, including the sealing of soil with impervious surfaces and the addition of anthropogenic materials such as construction rubble and waste materials. These anthropogenic influences alter the soil's ability to store carbon. However, despite the recent interest in urban soils, little is known about the impact of urbanisation on the long-term persistence of soil carbon, and in particular the stability of carbon in sealed soils. In this study, we investigate the functional pools of soil organic matter (SOM) in soils from urban greenspaces and under sealed surfaces. We separated particulate organic matter (POM) and mineral-associated organic matter (MAOM) pools to provide insights into soil carbon persistence and stability. We found that sealing negatively affected soil organic carbon storage across both functional SOM pools, illustrating the depletion of the persistent MAOM pool of organic carbon. In some instances, anthropogenic additions to sealed soils added legacy black carbon which increased the POM pool but did not contribute to increased organic carbon in the MAOM pool. Functional SOM patterns in urban greenspace soils highlighted the importance of these soils for persistent soil organic carbon storage with long residence times. However, anthropogenic additions to greenspace soils reduced N stocks, N in the MAOM pool, and dissolved organic carbon, highlighting the potential long-term negative impacts of urbanisation on ecological processes, nutrients and OC stores. Our findings highlight the consequences of both sealing and anthropogenic additions on long-term soil carbon persistence and the ecological processes occurring in sealed and urban greenspace soils.

4.1 Introduction

Soil organic matter (SOM) is an important soil property due to its contribution to soil carbon (C) storage and climate regulation, as well as supporting numerous other soil ecosystem services (Lorenz and Lal, 2015; Milne et al., 2015; Masciandaro et al., 2018). Urban soils have been increasingly studied for their potential to store large amounts of organic carbon (OC) and provide other ecosystem services associated with SOM (Pouyat et al., 2006; Qian and Follett, 2012; Edmondson et al., 2014a; Lorenz and Lal, 2015; Vasenev and Kuzyakov, 2018; Pouyat et al., 2020; O'Riordan et al., 2021a - chapter 3).

To date, few studies have considered urban soil OC across both urban greenspaces and under sealed surfaces such as roads and built surfaces (Edmondson et al., 2012; Cambou et al., 2018). Soil nitrogen (N) plays a key role in soil OC sequestration in many ecosystems due to its limitation on both primary productivity and microbial decomposition, therefore, it is necessary to consider N when investigating OC storage (Groenigen et al., 2006; Tipping et al., 2017; Averill and Waring, 2018; Davies et al., 2021; Rocci et al., 2021), as well as the influence of the urban environment on this relationship (Lorenz and Lal, 2009). Sealed surfaces often cover more than 50% of a city (Fuller and Gaston, 2009) and as urban areas are set to keep expanding, it is expected that sealing will become one of the key threats to soil sustainability (Seto et al., 2011; EU, 2012). When sealed surfaces are constructed, topsoil is removed before subsoil is compacted, road foundations are built and it is sealed with impermeable surfaces (Scalenghe and Marsan, 2009). In general, soil sealing reduces soil C and N storage (Raciti et al., 2012; Wei et al., 2014a; Wei et al., 2014b; Majidzadeh et al., 2017; Majidzadeh et al., 2018), with studies finding that sealing reduces soil C by between 37 - 68 % (Wei et al., 2014b; O'Riordan et al., 2021a - chapter 3). Sealing has also been shown to reduce microbial biomass and activity in sealed soils (Zhao et al., 2012; Wei et al., 2014a; Piotrowska-Długosz and Charzyński, 2015; Pereira et al., 2021). However, a small number of studies have challenged the assumption that sealing always leads to lower C stores. While soil C is lost during sealing as a result of topsoil removal, C stocks in the remaining soil are not always lower than greenspace soils (Edmondson et al., 2012; Cambou et al., 2018). When subsoil is taken into consideration, sealed soils can contribute notable soil C stores, and it has been suggested that sealing may isolate these from decomposition (Vasenev and Kuzyakov, 2018).

In urban areas, both greenspace and sealed soils are highly altered by human activities and their properties can be dominated by the addition of anthropogenic materials (Lehmann and Stahr, 2007; Pavao-Zuckerman, 2012). As a result of human activity and construction work, urban soils can also experience the mixing or burial of soil horizons (Weil and Brady, 2017; Herrmann et al., 2018) and the addition of anthropogenic materials into native soils (Vasenev et al., 2013; O'Riordan et al., 2021a). The effect of these additions to urban soils has been well studied in relation to human health and the increase in heavy metals and persistent organic pollutants (Li et al., 2018; Li et al., 2019; Brevik et al., 2020). However, additions can also contribute and alter C or N cycles and this has been less well-studied. Carbon and N can be added to urban soils from current or historical sources, such as atmospheric deposition (Tipping et al., 2017), brake ware (Grigoratos and Martini, 2015) or black C (pyrogenic C) which is a product of incomplete combustion of fossil fuels and biomass (Bird et al., 2015). Black C is ubiquitous in urban soils as result of historical soot emissions from heavy industry and power generation and can contribute more than 25% of the total urban soil OC pool (Novakov and Hansen, 2004; Edmondson et al., 2015; Schifman et al., 2018). Black C can contribute stable, recalcitrant forms of C to the soil which persist in soils despite not being associated with soil minerals (Marschner et al., 2008). Black C (in the form of biochar) has also been found to increase microbial biomass and activity due to its ability to alter soil pH, structure and nutrients (Zhang et al., 2018) and provide a labile C source for microbes (Cross and Sohi, 2011). Therefore, there is a need to consider the historical and cultural influence on urban soil carbon and the impacts of anthropogenic additions on soil functions (Vasenev et al., 2013; Ziter and Turner, 2018; Cambou et al., 2021). At present, we have little knowledge of the impacts of these additions on urban soil C and N storage and OC stability.

The persistence of organic matter (OM) in soil occurs due to chemical association with clay minerals, physical protection through aggregation, or biochemical stabilisation due to the recalcitrance of SOM compounds (Six et al., 2002). Current understanding is that SOM stability is determined by the accessibility of SOM to microbial decomposition and the soil environment, while the inherent recalcitrance of compounds is of less importance in controlling stabilisation (Elliott and Cambardella, 1991; Lützow et al., 2006; Marschner et al., 2008; Schmidt et al., 2011; Dungait et al., 2012; Hoffland et al., 2020; Lehmann et al., 2020). Recent frameworks have sought to separate SOM into functional pools to gain insights into SOM dynamics. These functional pools of particulate organic matter (POM) and mineral-associated organic matter (MAOM) can inform our understanding of SOM in the context of OC storage, OC persistence and soil functions (Janzen, 2006; Lopez-Sangil and Rovira, 2013; Trigalet et al., 2017; Cotrufo et al., 2019; Lavallee et al., 2020; Rocci et al., 2021). In the MAOM pool, protection by association with clay minerals arises due to the formation of organo-mineral complexes and is considered to be the most important mechanism for stabilising OC with long residence times (Mikutta et al., 2006; von Lützow et al., 2007; Kögel-Knabner et al., 2008; Poeplau et al., 2018). MAOM is largely made up of microbially-derived products and, as such, is a result of microbial activity (Kleber et al., 2015; Cotrufo et al., 2019). Alternatively, POM, made up largely of plant-derived material, can be free or occluded within macro-aggregates and while it may have some protection by aggregation, it is considered more accessible to microbes (Baldock and Skjemstad, 2000; Christensen, 2001; Six et al., 2001; von Lützow et al., 2007; Averill and Waring, 2018). However, its presence plays an important role in supporting SOM functions such as water holding, nutrient cycling and erosion resistance (Schmidt et al., 2011; Milne et al., 2015; Baveye et al., 2020).

Very few papers have considered the stability of OC in sealed soils (Wei et al., 2014b) and to our knowledge, no studies have yet considered the functional pools of SOM across both greenspace and sealed soils to understand OC persistence. In this study, we investigate the effects of sealing and anthropogenic additions on soil C stability, and use SOM functional pools to gain insights into OC persistence. Our research questions are: 1) What are the effects of sealing on urban soil OC and N persistence; and 2) what do anthropogenic additions to urban soils mean for soil C, N and OC persistence across sealed and greenspace soils? We hypothesise that (i) sealed soils will have less OC in the MAOM than greenspace soils and therefore less persistent OC; and (ii) soils with anthropogenic additions will have more OC in the MAOM and therefore more persistent OC in both sealed and greenspace soils. We analysed soils from greenspaces and from under sealed surfaces across the historic city of Manchester, UK, and determined whether the soils were relatively undisturbed or contained anthropogenic additions as a result of human activity. We used SOM functional pools to understand soil C and N storage and gain insights into OC persistence. We separated functional pools of SOM into POM and MAOM using physical fractionation: POM (2000-50 μm fraction) and MAOM (<50 μm fraction), and measured C and N content and dissolved carbon in the supernatant.

This study addresses a gap in our understanding of the complexity of soil C stability across the urban landscape. It has implications for the long-term storage dynamics of sealed soils and the consequences of construction activities, as well as the implications of anthropogenic additions to greenspace and sealed soils in the context of C storage and persistence.

4.2 Methods

4.2.1 Soil Sampling and Categorisation

Soils were sampled from Manchester, UK, as described by O'Riordan et al. (2021a - chapter 3). Briefly, sealed soils were collected from roadworks where work had recently exposed the sealed soil, and greenspace soils were collected from the nearest greenspace, park or roadside to the sealed sample and within grassed lawn areas. Both soils were sampled to 10 cm depth, in sealed soil this was the top 10 cm of soil underneath the construction material; in greenspace soil this was the top 10 cm under the turf and litter layer. In sealed soils, the profile and depth of available soil under construction materials varied across sites, though most samples were collected between 60-80 cm depth. In total, 68 sites were sampled with 36 of these sealed sites and 32 greenspace sites. These were then categorised according to the presence of anthropogenic materials (as described below in 4.2.1.1) with the following sample numbers: greenspace anthropogenic (GSA; n=11), greenspace undisturbed (GSU; n=21), sealed anthropogenic (SA; n=14), and sealed undisturbed (SU; n=22) soils (figure 4.1).

4.2.1.1 Soil categorisation

The World Reference Base for Soil Resources (WRB) defines Technosols as either containing a large volume of artefacts, having an impermeable geomembrane or having a 'technic hard material' such as concrete, asphalt or stone at the soil surface (FAO, 2015). The sealed soils sampled in this study are therefore considered Technosols due to the continuous hard surface. In addition, we identified human-made artefacts in both the greenspace and sealed soils, such as concrete, bricks, charcoal, glass and plastic; however not all soils contained these artefacts and some were relatively undisturbed with few additions of human-made materials. These additions have the potential to greatly influence the mineral, carbon and nutrient status of the soil, and therefore, the samples were categorised into four types. Soils that were relatively undisturbed (other than by sealing) were classed as either sealed undisturbed (SU) or greenspace undisturbed (GSU); and soils with notable additions of anthropogenic materials were classed as sealed anthropogenic (SA) or greenspace anthropogenic (GSA). To distinguish between these types we used the proportion of coarse material in the soil as a proxy for anthropogenic additions, as described by O'Riordan et al. (2021a - chapter 3). Briefly, wet sieving was undertaken on subsamples of all samples and the proportion of material in the >200 μm fraction was used to determine the amount of anthropogenic additions. Soils with visible additions exhibited 40 % or greater subsample mass in the >200 μm fraction; thus subsamples that exhibited this pattern were classed as anthropogenic soils, and those with less than 40 % mass were classed as undisturbed, across both sealed and greenspace soils. The fragmentation of materials into smaller fractions made it inaccurate to measure artefact mass alone; while this method served to describe the samples well and allow a comparison between anthropogenic and undisturbed soils.

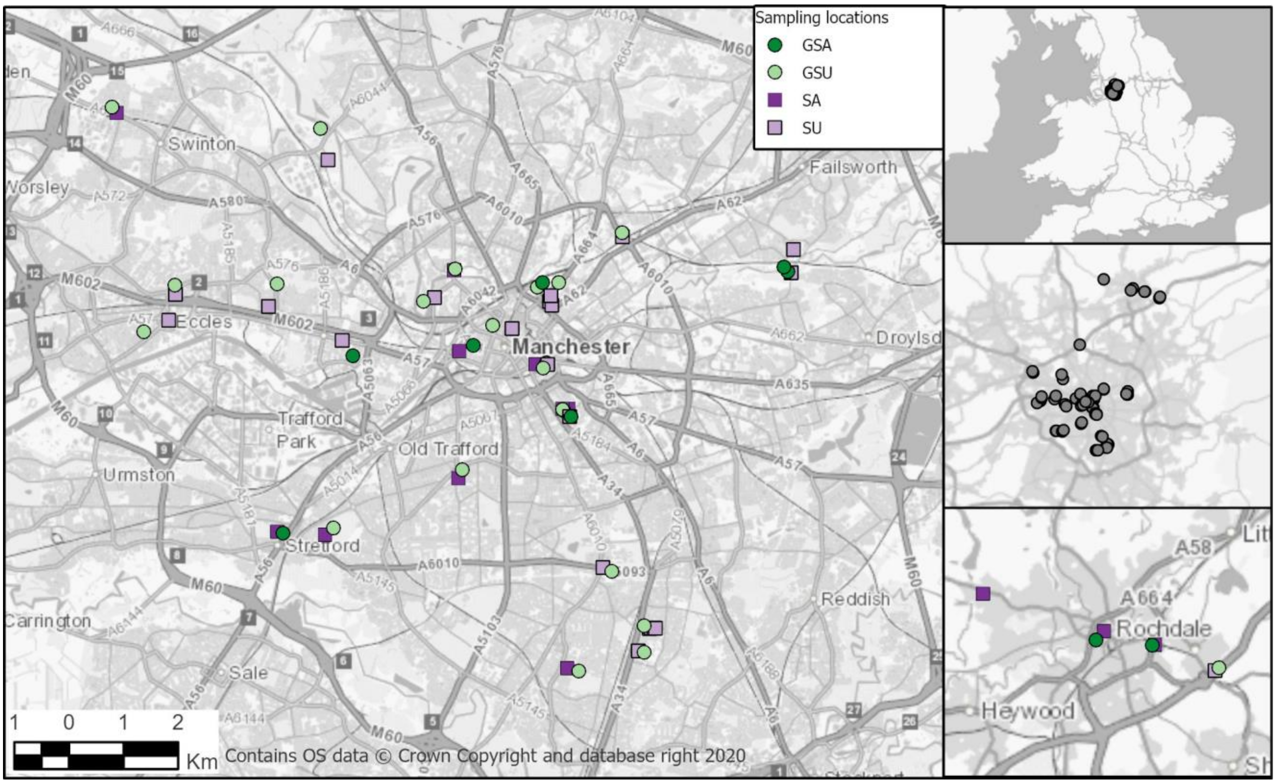

Figure 4.1: Map showing the sampling locations across the Greater Manchester region. Soil categories are greenspace anthropogenic (GSA; n=11), greenspace undisturbed (GSU; n=21), sealed anthropogenic (SA; n=14), and sealed undisturbed (SU; n=22). Sealed soils are represented by squares and greenspace soils are represented by circles; depth of colour indicates whether soils were undisturbed or anthropogenic.

4.2.2 *Soil Analyses*

4.2.2.1 Physical fractionation

Physical fractionation to separate size fractions was undertaken following the method by Lopez-Sangil and Rovira (2013). Duplicate dried subsamples (15 g each) were weighed into 50 ml corning tubes with two glass marbles and 30 ml MQ water in each. Tubes were placed in an end-over shaker (18 rmp) for 1 hour. The glass marbles were removed, the soil-water mixture was transferred to an extraction bottle and was made up to 150 ml with MQ water.

Sonication was undertaken to disperse large secondary aggregates from primary aggregates and particles (Roscoe et al., 2000) using a Sonics 130 Watt ultrasonic processor with 13 mm probe. The soil-water mixture was kept in an ice bath to prevent heating, and the probe was inserted 15 mm into the mixture. The amount of energy delivered to the mixture was determined using the equation proposed by Christensen (1985) (equation 1), where E_a is the applied energy (J ml^{-1}), P_c is the power (W), t_s is the time (seconds) and V is volume of suspension (ml).

Equation 1 $E_a = P_c\, t_s / V$

The sonicator was used at 70% amplitude which corresponds to an output of approximately 39 W. Using the equation above, the mixture was sonicated for 1,000 seconds to reach an energy of 260 J ml^{-1}, a threshold identified to disperse macroaggregates but prevent redistribution of C into smaller fractions (Roscoe et al., 2000).

Immediately after sonication, the mixture was separated by wet sieving into size fractions of >2000, 2000-200, 200-50, 50-20 and <20 μm (coarse fraction, coarse sand, fine sand, coarse silt and clay, respectively). The <20 μm fraction in the receiving sieve was made up to 1 litre volume with MQ water in a Buchner flask, mixed with 8.7 g of K_2SO_4 as a flocculent and left for 24 hours. After sedimentation, the supernatant was siphoned off and the remaining portion was centrifuged for 15 mins at 2500 rpm. The liquid was siphoned off and the pellet retained to form the <20 μm fraction. All fractions were dried at 60 °C for 24 hours then weighed. The supernatant was used for dissolved OC and inorganic carbon (IC) analysis.

4.2.2.2 *Inorganic carbon removal*

Inorganic carbon (carbonate) was removed from the fractions using HCl gas fumigation, as described by Harris et al. (2001) and amended by Ramnarine et al. (2011). The acid fumigation method was chosen to prevent the loss of water-soluble C that can arise due to the use of liquid acid (Harris et al., 2001). Glass beakers were used rather than silver boats to prevent the loss or spillage or samples during the procedure, as suggested by Ramnarine et al. (2011).

The oven dried fractions were ground using a ball-mill. Subsamples of 300 mg were placed in 5 ml glass beakers and moistened with 150 µl MQ water. The beakers were placed in a glass desiccator together with a beaker of 100ml of 12 M HCl acid; this was vacuum sealed and left to fumigate for 72 hours. After this, the HCl beaker was removed and vacuum evacuation was repeated 10 times to remove HCl vapour. The fractions were dried at 105 °C for 24 hours.

During HCl fumigation, carbonate is converted to CO_2 and $^-Cl_2$ ions. The $^-Cl_2$ ions weigh more than the carbonate removed so the final sample has a heavier mass after fumigation and, thus, the remaining organic C is diluted within the new heavier sample mass. To account for this, the change in mass from before and after fumigation was recorded to obtain a correction factor for each fraction. This correction factor was used to calculate the actual mass of soil used for CN analysis for each fraction.

4.2.2.3 CN and functional SOM pool analysis

Size fractions were analysed for OC and total N concentration by combustion oxidation (Vario EL Cube CHNS Elemental Analyser, Elementar, Germany). Organic C and total N content in each fraction were corrected for fraction mass within the bulk soil. Overall soil OC and total N content were determined as the sum of OC and total N measured across all size fractions. Overall soil OC and N stocks were calculated using bulk density values to 10 cm depth (see chapter 3) and OC and N contents, as per the guidance from FAO (2018). Dissolved OC and IC analysis was undertaken on the supernatant removed during fractionation using a Total Organic Carbon (TOC) analyser (Shimadzu TOC-L CPN TN). Dissolved OC is determined as total dissolved C minus dissolved IC.

Functional pools of SOM were separated into particulate organic matter (POM) and mineral-associated organic matter (MAOM) pools, as per recommendations by

Lavallee et al. (2020) and Cotrufo et al. (2019). POM was determined as particles in the 2000-50 μm size fractions; and MAOM as particles in the <50 μm size fractions.

4.2.3 Statistical analysis

The distance between sealed and greenspace samples varied and as a result they were not treated as paired samples. Data distribution was checked for normality using the Shapiro-Wilk test. Where data was not normally distributed it was transformed using either log10 or square root transformation. Sealing and the addition of anthropogenic materials were treated as two factors, and therefore, a two-way ANOVA was performed to test for the effects of sealing, additions, and the interaction between these two factors. A Tukey post hoc test was then used to understand the interaction further and identify where the differences lay between soil categories. Where transformation did not result in data with normal distribution, ANOVA results were checked for consistency against results from the non-parametric test equivalent, the Scheirer-Ray-Hare test and Dunn post hoc test, performed using the original data. Where results from the ANOVA and Scheirer-Ray-Hare tests differed, the Scheirer-Ray-Hare results were reported. This applied only to the overall OC stock variable. All statistical analyses were undertaken in R version 4.1.0.

4.3 Results

4.3.1 Overall soil OC and N storage

In comparing undisturbed sealed and greenspace soils (SU and GSU) we found that sealed soils had significantly lower OC content ($p < 0.0001$) and OC stock ($p < 0.0001$) in comparison to greenspace soils (figure 4.2 a, b). Sealed soils also had lower total N content ($p < 0.0001$) and total N stock ($p < 0.0001$) compared to greenspace soils (figure 4.2 c, d).

Anthropogenic additions appeared to affect soil OC differently depending on whether soils were sealed or not. There was a significant interaction between sealing and anthropogenic additions on OC content ($F_{1,64} = 16.648$, $p < 0.001$), OC stock ($H_{1,64} = 8.441$, $p < 0.01$), and total N content ($F_{1,64} = 4.842$, $p < 0.05$), indicating that the effect of additions varied according to sealing status. Soil OC was greater in sealed soils with additions (SA) to the extent that OC content and OC stocks were of a similar order to those found in greenspace (GSA) soils ($p = 0.252$ and $p = 0.645$ respectively). However, the opposite was found in greenspace soils with additions (GSA) which had lower OC content and OC stock than undisturbed greenspace soils (GSU), though this effect was not significant.

Soil N appeared be consistently lower in soils with additions, across both sealed and greenspace soils. Sealed soils with additions (SA) exhibited significantly lower N stock than SU soils ($p < 0.01$); while greenspace soils with additions (GSA) had significantly lower N content ($p < 0.01$) and N stock ($p < 0.01$) than GSU soils (figure 4.2 c, d).

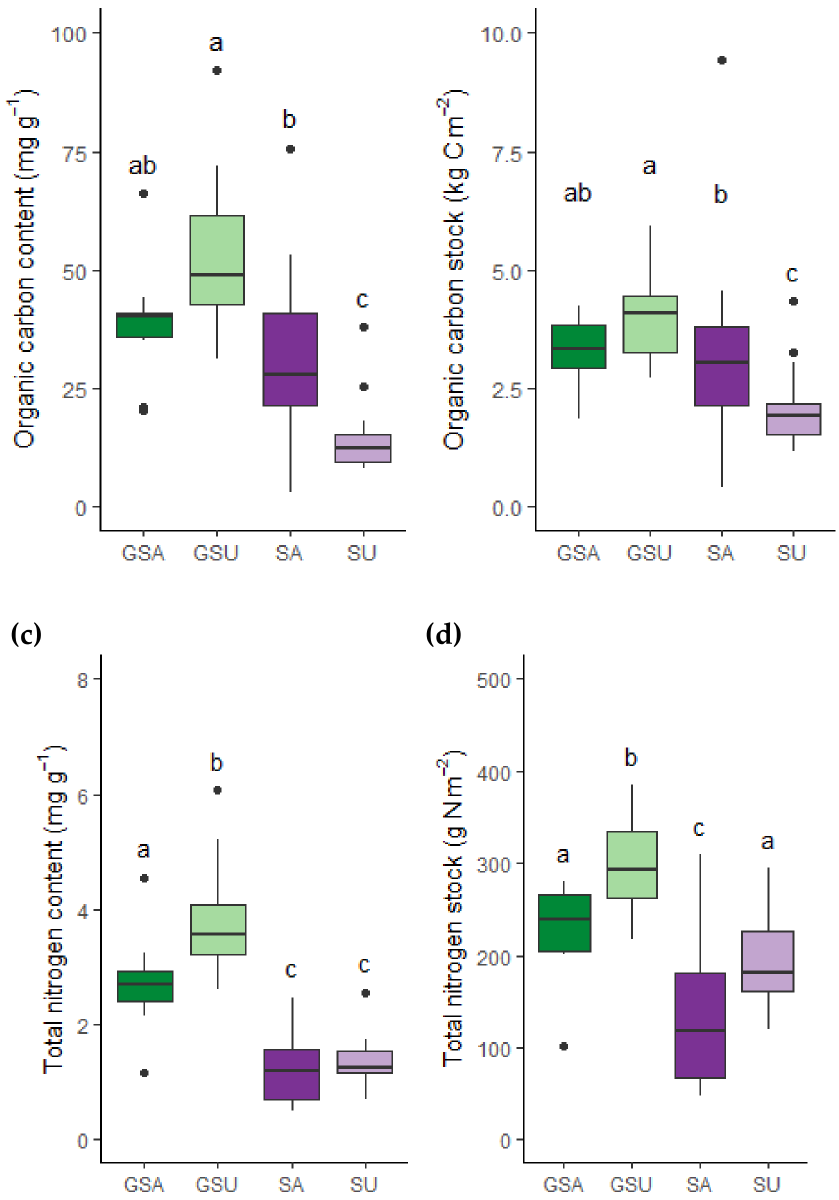

Figure 4.2: Overall soil data, showing (a) organic carbon content (mg g^{-1}), (b) organic carbon stock (kg m^{-2}), (c) total nitrogen content (mg g^{-1}), (d) total nitrogen stock (g m^{-2}). Soil categories are greenspace anthropogenic (GSA), greenspace undisturbed (GSU), sealed anthropogenic (SA), and sealed undisturbed (SU). Stocks are calculated for 0-10cm at the depth sampled for each soil type. Different letters indicate a significant difference at p = 0.05. The box represents the upper and lower quartiles, whiskers show upper and lower most values, and horizontal lines show the median. Circled data points show outliers at 1.5 times the interquartile range.

4.3.2 *OC and N in size fractions and functional SOM pools*

Across all size fractions, the clay fraction (<20 µm) generally exhibited the highest OC and N contents (figure 4.3 a, b). In sealed undisturbed soil (SU), there was notably lower OC and N content in the larger size fractions compared to greenspace (GSU) soil, while the clay fraction (<20 µm) remained the largest store of OC and N content. The pattern of OC and N storage across fraction sizes was similar in both SU and GSU soils, only on a much smaller scale in the SU soil.

Anthropogenic additions changed the distribution of OC and N content within the size fractions. In sealed anthropogenic (SA) soils, there was greater storage of OC and N in the larger size fractions. This was most prominently seen in OC content which was highest in the coarse sand fraction (2000-200 µm), while N content in this fraction was second highest after the clay fraction (<20 µm). Conversely, in greenspace anthropogenic (GSA) soils, there was notably less OC and N content in the fine sand fraction (200-50 µm), as well as less OC and N in the coarse silt (50-20 µm) and clay (<20 µm) fractions.

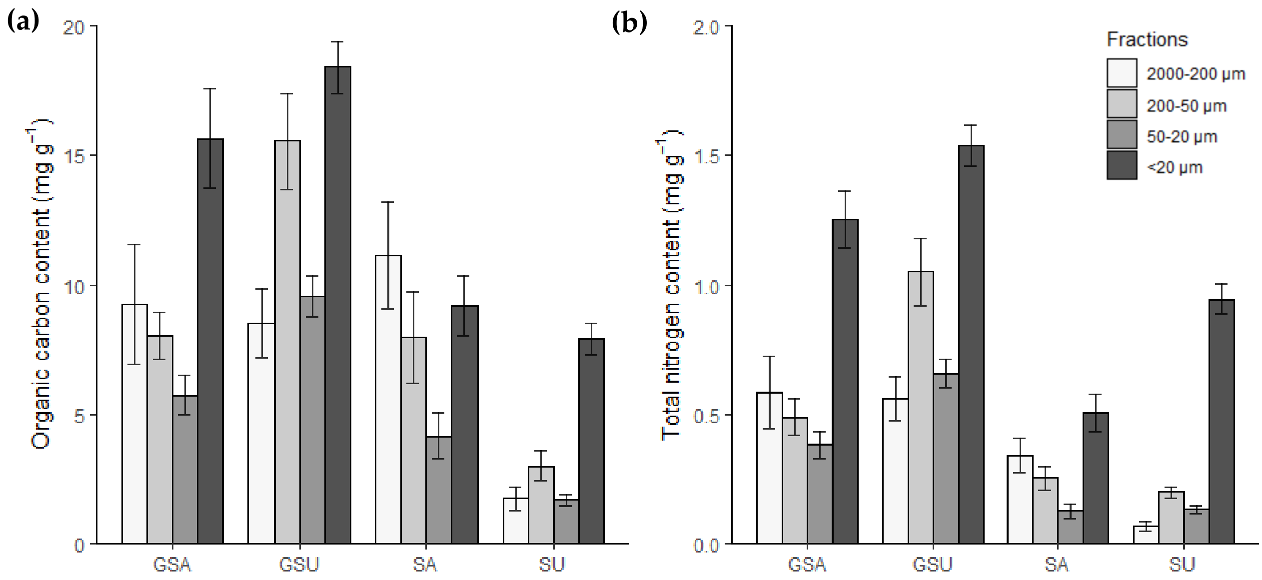

Figure 4.3: Size fractions showing (a) organic carbon content (mean + SE in mg g^{-1}); (b) total nitrogen content (mean + SE in mg g^{-1}). Soil categories are greenspace anthropogenic (GSA), greenspace undisturbed (GSU), sealed anthropogenic (SA), and sealed undisturbed (SU).

To consider the functional pools of SOM, we studied OC and N stored in particulate organic matter (POM) and mineral-associated organic matter (MAOM). In comparing undisturbed sealed and greenspace soils (SU and GSU), we found that sealed soils had significantly lower OC and N contents in the POM pool (both $p < 0.0001$) and the MOAM pool (both $p < 0.0001$) compared to greenspace soils (figure 4.4 a, c). The proportion of OC and N in POM was significantly lower in SU soils than GSU soils; thus, the proportion of OC and N in MAOM was significantly higher in SU soils than GSU soils (figure 4.4 b, d).

Anthropogenic additions affected the POM and MAOM pools differently. In the POM pool, anthropogenic sealed (SA) soils had significantly greater POM OC and N content than SU soils ($p < 0.001$ and $p = 0.039$ for OC and N respectively). This POM OC content was as high as that found in greenspace (GSU and GSA) soils ($p = 0.202$ and $p = 0.890$). This led to a larger proportion of OC and N to be stored in POM than in MAOM for SA soils (figure 4.4 b, d). Additions to greenspace soils appeared to have the opposite effect on POM, with POM OC and N contents in GSA soils slightly lower than in GSU soils, although this was not statistically significant (figure 4.4 a, c).

In the MAOM pool, additions did not have any significant effect on MAOM OC content in the sealed or greenspace soils (figure 4.4 a). However, MAOM N content was significantly lower in both sealed (SA) and greenspace (GSA) anthropogenic soils compared to undisturbed (SU and GSU) soils ($p = 0.001$ in sealed soils; $p < 0.001$ in greenspace soils) (figure 4.4 c). Greenspace soils had a larger proportion of OC and N stored in the MAOM pool compared to the POM pool, whether there were additions or not.

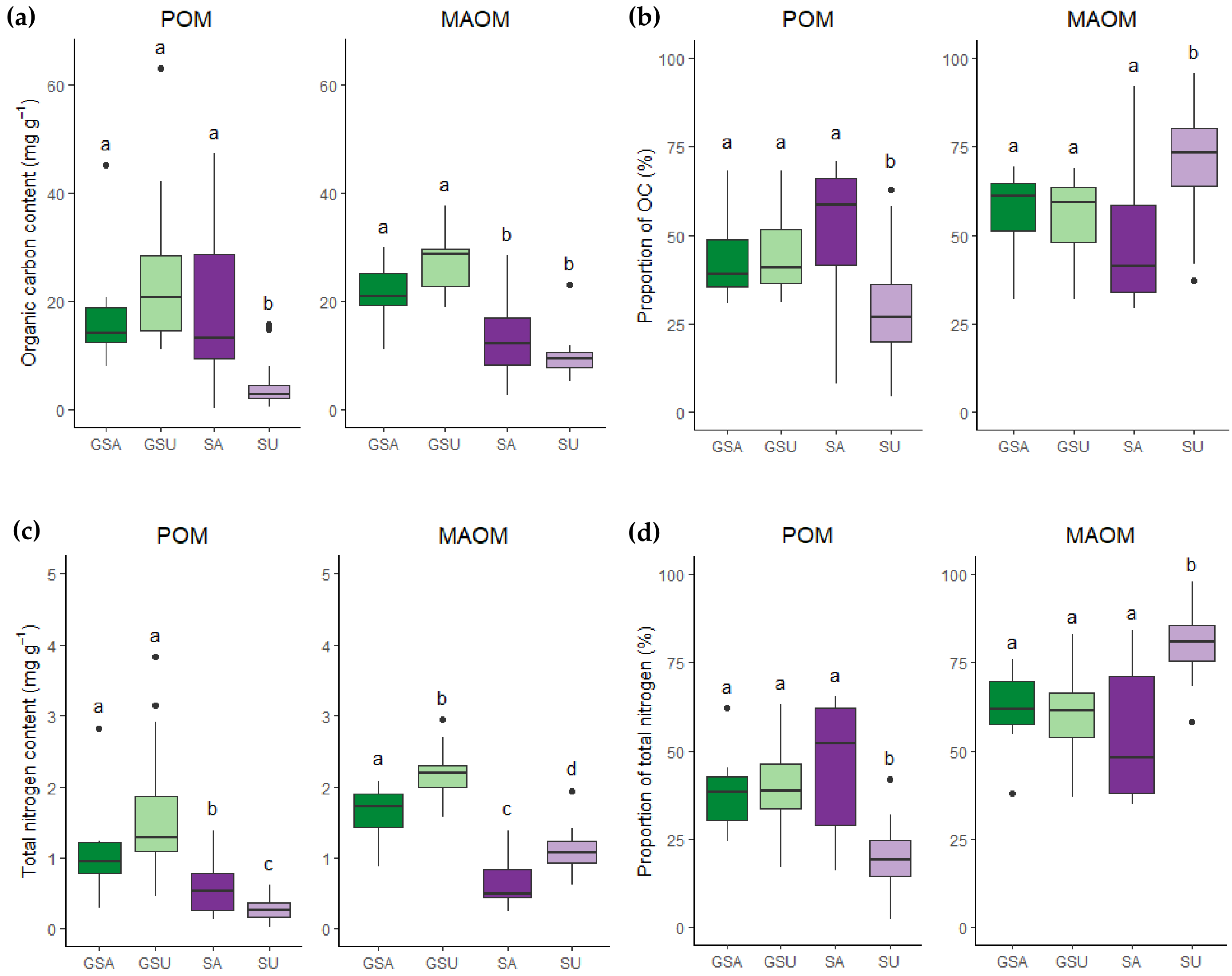

Figure 4.4: Differences in POM and MAOM showing (a) organic carbon content (mg g^{-1}); (b) proportion of organic carbon (%); (c) total nitrogen content (mg g^{-1}); (d) proportion of total nitrogen (%). Soil categories are greenspace anthropogenic (GSA), greenspace undisturbed (GSU), sealed anthropogenic (SA), and sealed undisturbed (SU). The box represents the upper and lower quartiles, whiskers show upper and lower most values, and horizontal lines show the median. Circled data points show outliers at 1.5 times the interquartile range.

Dissolved OC and inorganic C (IC) were also measured in the supernatant recovered during physical fractionation. The dissolved OC data indicated a significant interaction between sealing and additions, suggesting that additions affected dissolved OC differently in sealed and greenspace soils ($F_{1,64} = 9.726$, $p < 0.01$; figure 4.5 a). In comparing undisturbed soils, sealed soils (SU) had significantly lower dissolved OC content than greenspace (GSU) soils ($p < 0.0001$). Additions to sealed soils did not affect dissolved OC content. However, in greenspace anthropogenic (GSA) soils, dissolved OC content was significantly lower than in undisturbed GSU soils ($p < 0.01$).

Dissolved inorganic carbon (IC) data illustrated that both sealing and additions could lead to greater dissolved IC content (figure 4.5 b). In comparing undisturbed soils, sealed soils (SU) had significantly greater dissolved IC than greenspace (GSU) soils ($p < 0.05$). In sealed soils, additions did not affect dissolved IC content and both SU and SA soils had similar high dissolved IC content. In greenspace anthropogenic (GSA) soils there was significantly greater dissolved IC content than in GSU soils ($p < 0.05$). Greenspace soils appeared to be more affected by the additions in terms of dissolved OC and IC content.

Soil pH was significantly higher in sealed (SU) soil than in GSU soil ($p < 0.0001$; figure 4.5 c). In sealed soils, the additions did not alter the pH and both SU and SA soils had high soil pH. In greenspace anthropogenic (GSA) soils the pH was significantly higher than in GSU soils ($p < 0.001$).

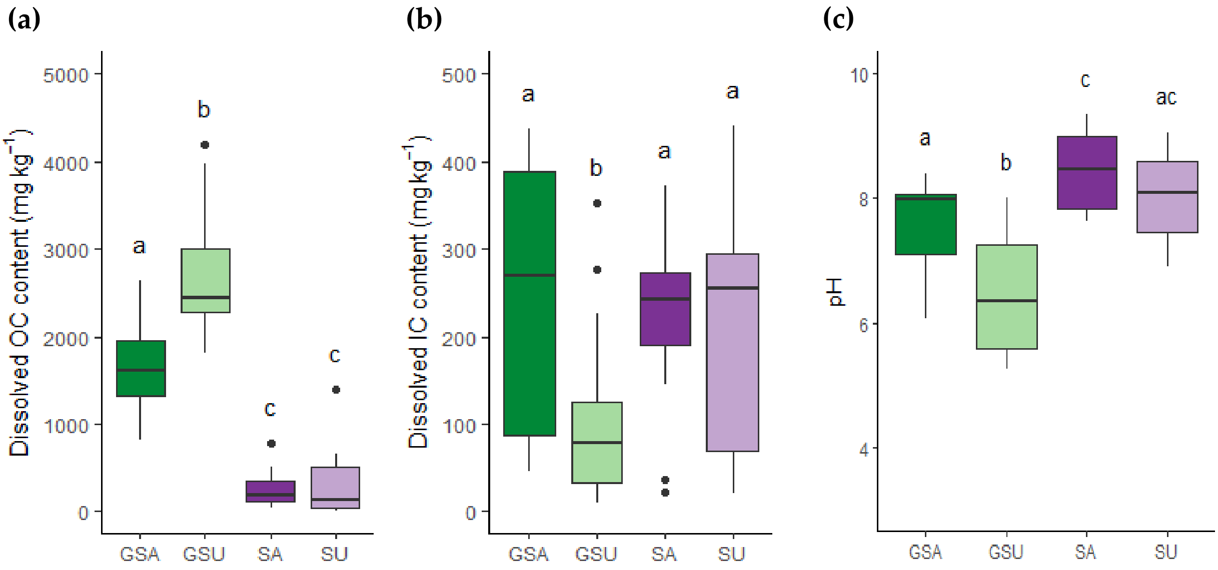

Figure 4.5: Dissolved carbon recovered from supernatant and pH, showing (a) dissolved organic carbon (mg kg^{-1}); (b) dissolved inorganic carbon (mg kg^{-1}); and (c) soil pH. Soil categories are greenspace anthropogenic (GSA), greenspace undisturbed (GSU), sealed anthropogenic (SA), and sealed undisturbed (SU). The box represents the upper and lower quartiles, whiskers show upper and lower most values, and horizontal lines show the median. Circled data points show outliers at 1.5 times the interquartile range.

4.3.3 *CN Ratio*

In comparing undisturbed sealed and greenspace soils (SU and GSU), we found no significant difference in the CN ratio of the POM pool, though there was much greater variation in CN ratio. In the MAOM pool, however, the CN ratio was significantly lower in sealed soils (figure 4.6 a). In addition, there was much greater variation in the POM CN ratio in the sealed soil.

There was a significant interaction between sealing and additions on POM CN ratio ($F_{1,64} = 4.071$, $p < 0.05$), and MAOM CN ratio ($F_{1,64} = 34.488$, $p < 0.0001$), indicating that additions affected sealed and greenspace soils differently. In sealed anthropogenic (SA) soils, the CN ratio was much greater than in SU soils for the POM pool and MAOM pool (figure 4.6 a). In particular, the MAOM pool CN ratio was much wider in range than the narrow CN ratio seen in the SU soil. In greenspace soils, additions did not significantly alter the CN ratio in either the POM pool or MAOM pool.

These patterns were also observed in correlations between overall soil CN ratio and POM and MAOM CN ratios (Figure 4.6 b, c). As the overall CN ratio increases, the POM CN ratio increases most in the SA soil followed by the SU soil, while both greenspace soils exhibit less variation in CN ratio (figure 4.6 b). When correlating overall CN ratio with the MAOM CN ratio, SU soil exhibits a similar pattern to the two greenspace soils, with lower and a much narrower range of MAOM CN ratios than the SA soil (figure 4.6 c).

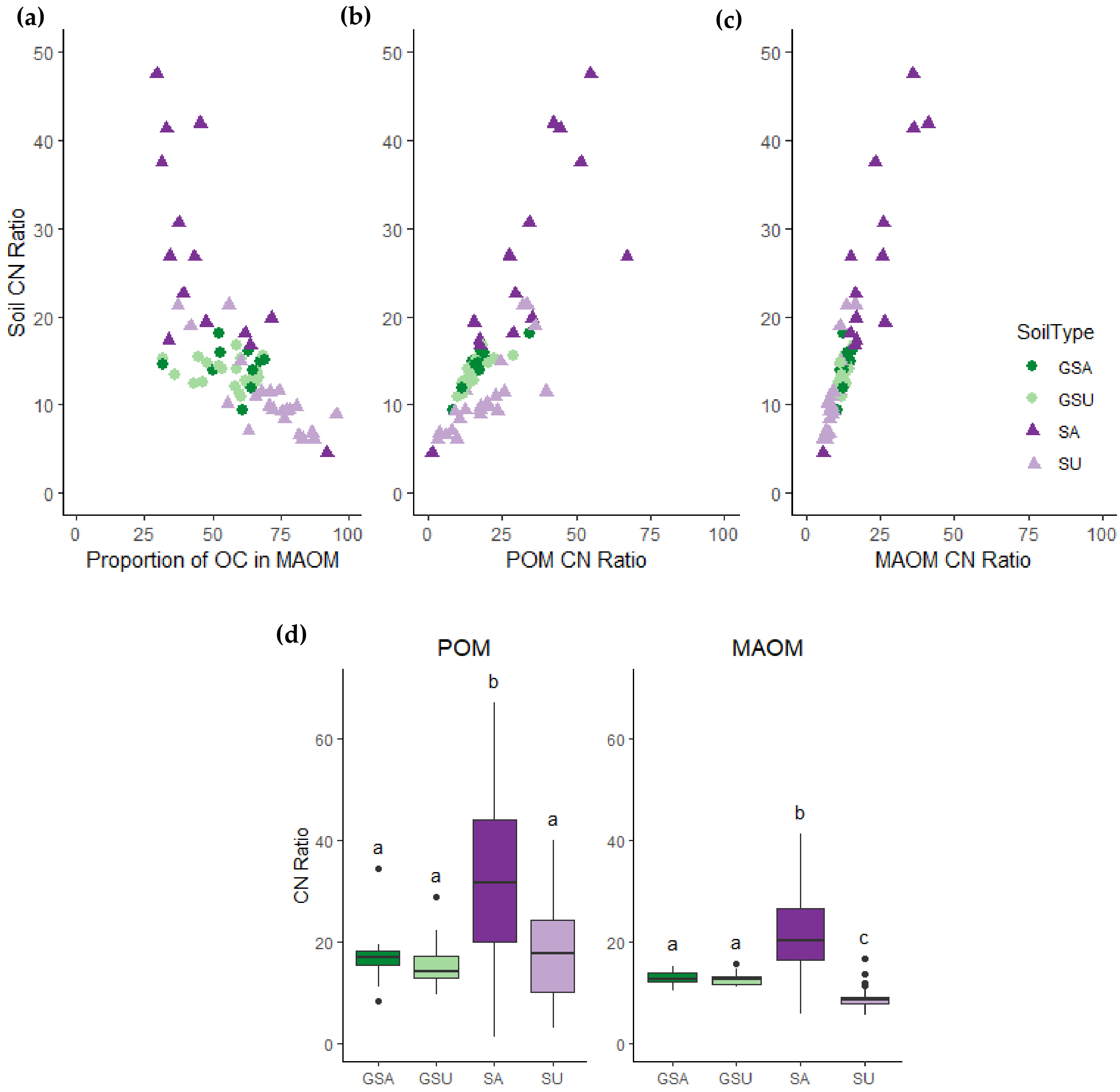

Figure 4.6: a-c show overall soil CN ratio plotted against (a) proportion of OC in the MAOM pool (%); (b) POM CN ratio; (c) MAOM CN ratio; (d) shows CN ratio in POM and MAOM pools. Soil categories are greenspace anthropogenic (GSA), greenspace undisturbed (GSU), sealed anthropogenic (SA), and sealed undisturbed (SU). The box represents the upper and lower quartiles, whiskers show upper and lower most values, and horizontal lines show the median. Circled data points show outliers at 1.5 times the interquartile range.

4.4 Discussion

4.4.1 Sealed soils have lower accessible and stable SOM

Sealed soils (SU) that were relatively undisturbed by additions of anthropogenic material had significantly less OC and N stock compared to greenspace (GSU) soils. These results are to be expected and follow the pattern seen in the majority of previous studies into sealed soils, where soil OC content is typically reduced as a result of soil sealing (Raciti et al., 2012; Wei et al., 2014a; Wei et al., 2014b; Piotrowska-Długosz and Charzyński, 2015; Yan et al., 2015; Majidzadeh et al., 2017; O'Riordan et al., 2021a; Pereira et al., 2021).

With respect to the persistence of OC, which is the novel focus of this work, we hypothesised (i) that sealed soils would have less mineral-associated OC than greenspace soils and therefore less persistent OC. We found that SU soils had significantly lower OC and N content than GSU soils in both the POM and MOAM pools (figure 4.4 a, c). Our results illustrate that sealing depletes both the accessible POM OC store, as well as the long-term persistent MAOM OC store. Therefore, we accept hypothesis i, as the results illustrate that there is less persistent OC in sealed soils than in greenspace soils.

The reduction in POM OC may be due to a number of reasons, including the removal of topsoil during sealing, a lack of OM inputs, and the continued loss of SOM through decomposition. The removal of topsoil during sealing would lead to the loss of POM and, as a result, the soil studied may be more similar to a subsoil in terms of POM content. The fractionation data illustrated that SU and GSU soils had a similar pattern of OC and N content across size fractions, though on a smaller scale for SU soils (figure 4.3), suggesting that similar processes of OC and N storage may be occurring on a smaller scale in SU soil. In addition, the lack of plant growth would lead to a lack of OM inputs from roots and litter. As POM is primarily made up of plant-derived organic matter (Six et al., 2001), the lack of plant growth would prevent replenishment of the POM pool in sealed soils. It is also possible that POM has been lost through decomposition following sealing such that the POM pool has been largely diminished. Soils with sufficient POM would be expected to exhibit measurable dissolved OC, present as a result of being easily dissolved in soil water (Kalbitz et al., 2000). We found that SU soils had nearly ten times less dissolved OC than GSU soils, indicating that labile OC was depleted in these sealed soils. This was also observed by Wei et al. (2014b) who found that sealed soils were depleted of readily decomposable OC.

Both POM and dissolved OC are easily accessible to soil fauna and provide an energy source to facilitate decomposition (Wu et al., 2018; Lavallee et al., 2020). With this no longer readily available it is likely that decomposition is prevented, or at least severely reduced, in sealed soils. It is also likely that as microbial activity is reduced, microbial biomass and necromass will not accumulate. Previous studies have illustrated that sealing reduces soil microbial biomass C and N, microbial functional diversity, soil respiration, N transformation and enzyme activity, as well as altering the bacterial:fungal ratio, all of which indicate that microbial activity is lower in sealed soils (Zhao et al., 2012; Wei et al., 2013; Wei et al., 2014a; Wei et al., 2014b; Piotrowska-Długosz and Charzyński, 2015; Pereira et al., 2021). It has been suggested that OC in sealed soils has weaker decomposability and a lower turnover rate (Wei et al., 2014b). However, it would appear from the studies of microbial activity that the OC is not simply more stable, but that microbial biomass, activity and mineralisation are all significantly reduced. Therefore, there is the possibility that once POM is depleted, microbial biomass and activity declines in sealed soils.

The MAOM pool is largely made up of microbially-derived products which bind to soil minerals, giving them greater protection from microbial attack and allowing OC and N within MAOM to persist with long residence times (Kögel-Knabner et al., 2008). In sealed soils, it is likely that lower microbial activity leads to a lack of microbially-derived products to build up the MAOM pool. In addition, the lower MAOM OC and N contents in sealed soils suggest that some of the original MAOM, present prior to sealing, may have been lost since sealing occurred. Despite being thought of as stable, MAOM is not completely permanent and can be lost due to decomposition in some situations (Dynarski et al., 2020). Therefore, the low MAOM OC and N levels may be a result of losses of original MOAM after sealing occurred, a lack of replenishment due to low microbial activity, or both of these processes. Further studies into the processes occurring in sealed soils would build greater understanding of the losses of both POM and MAOM, and would potentially enable better management of urban soils for OC storage in construction.

4.4.2 Anthropogenic additions to sealed soils add black OC but not microbial OC

We set out to test hypothesis ii in sealed soils: that soils with anthropogenic additions will have more mineral-associated OC and therefore more persistent OC. In sealed anthropogenic (SA) soils, overall soil OC was much greater than in sealed undisturbed (SU) soils. However, much of this OC was in the POM pool, while OC in the MAOM pool was no greater than in SU soils. Therefore, we reject hypothesis ii for SA soils, as despite the higher OC in the POM pool, there is still less OC in the MAOM pool and less persistent OC than in greenspace soils.

Fractionation showed that the high OC in SA soil was most prominent in the largest coarse sand fraction (2000-200 μm), illustrating that the additions remained mostly in this fraction and contributed to its high OC. This resulted in the SA soil POM pool having significantly greater OC than in SU soils, such that OC was equivalent to POM OC in greenspaces. The high OC was likely due to black C present in the additions rather than plant-based POM. Black C (pyrogenic C) was commonly used in road foundations during the 18-19th century in the UK (and other countries such as the USA) as charcoal was a waste product from industry at that time (MacBride, 2013). Coal fly ash has also been used extensively in UK road construction as fill or converted into aggregate (Wainwright and Cresswell, 2001). The high OC in SA soils was not accompanied by equally high N content, and overall N was no greater in SA soil than in SU soil. Though there was greater POM N in the SA soil, it was not on the same scale as the increase in POM OC. This indicates further that in SA soils the high OC was a result of black C additions, which do not elevate soil N.

Black C is an important pool of soil OC due to its assumed long residence time within the soil (Bird et al., 2015; Abney and Berhe, 2018), though our understanding of its contribution to urban soils is limited (Edmondson et al., 2015). Black C is promoted, as biochar, as a means to increase agricultural soil OC content (Smith, 2016; Gelardi and Parikh, 2021); however, research into its effectiveness for OC sequestration is mixed (Wardle et al., 2008; Liang et al., 2010; Major et al., 2010; Liu et al., 2018; Crispo et al., 2021). It is thought that black C can be stabilised through soil burial as this reduces exposure to microbes and oxygen, reducing decomposition (Abney and Berhe, 2018). In the context of sealed soils, the burial of black C may serve as a mechanism for its stabilisation. Large stores of black C under sealed surfaces have been observed as a legacy carbon store in Manchester, UK, an area with a heavily industrial past (O'Riordan et al., 2021a). However, recent studies have illustrated that black C can be decomposed more quickly than previously thought, on the scale of months to years (Hilscher et al., 2009; Nguyen et al., 2010; Zimmerman, 2010; Bird et al., 2015). Therefore, it is thought that the persistence of black C in soil is a property of the ecosystem and soil conditions (Schmidt et al., 2011). This has implications for the redevelopment of cities and roads. Whilst roads remain intact it's likely this black C will remain stable and persist in the soil; however, during urban redevelopment or road and utility repairs there is the risk of exposing legacy black C stores, leading to its loss either through removal or decomposition.

By contrast, the OC in the MAOM pool in SA soils was no greater than in SU soils. In addition, there was very little dissolved OC content in SA soils. We suggest that black C did not contribute to microbial activity in these soils as microbes would have preferentially decomposed any labile OC (Nykvist, 1963) and contributed to a larger microbially-derived MAOM OC pool. Black C (or biochar) can provide a surface for adsorption of dissolved OC (Pietikäinen et al., 2000; Ukalska-Jaruga et al., 2019), and has been found to increase microbial biomass and activity due to its nutrients, structural properties and ability to alter the soil pH (Zhang et al., 2018). However, in this study, black C made no difference to MAOM OC storage in sealed soils. As the MAOM pool is usually microbially-derived, it is likely that the resource-limited environment of sealed soils prevented microbial activity, despite the presence of black C. The CN ratio of the MAOM pool was higher and wider in range in SA soils. This reflects the lower N in these soils, which indicates that biological activity was not occurring and did not lead to the N-rich OM microbial products usually found in the MAOM pool. This suggests that black C did not contribute to microbial activity and microbial OM storage. In addition, microbes have a constrained CN ratio within which they can function, with bacteria preferring a CN ratio of 3 to 5 and fungi preferring 4.5 to 15 (Paul, 2014; Cotrufo et al., 2019). The high CN ratio of the SA soil MAOM pool exceeds these preferred ratios, further indicating that the MAOM OC was likely not microbially-derived. Biochar is increasingly being used in urban soils to improve soil quality (Scharenbroch et al., 2013; Somerville et al., 2020). Therefore, this study has implications for the use of biochar in urban soils, as while it may contribute to OC storage as POM, if used in sealed soils it may not contribute to microbial activity or long-term microbial OC storage. It also highlights its vulnerability to loss should it be removed during roadworks and redevelopment. As interest in urban soil C storage increases, further research into the effects of biochar in urban soils, in both sealed and greenspace soils, would be beneficial.

4.4.3 Anthropogenic additions to greenspace soils reduce N stores and labile OC

We set out to test hypothesis ii in greenspace soils: that soils with anthropogenic additions will have more mineral-associated OC and therefore more persistent OC. In greenspace anthropogenic (GSA) soils, the overall soil OC content was lower than in undisturbed greenspace (GSU) soils, though this was not statistically significant. In addition, there was significantly less overall soil N content and N stock in GSA soil. Fractionation revealed no difference in MAOM OC between GSA and GSU soils, while there was significantly less MAOM N and less dissolved OC in the GSA soils.

Therefore, we reject hypothesis ii for GSA soils as, despite the additions, there is no increase in persistent OC.

The lower soil N and OC content in GSA soils were seen most clearly in the fine sand fraction (200-50 μm), as well as in the coarse silt (50-20 μm) and clay (<20 μm) fractions. This resulted in significantly lower N in the MAOM pool in GSA soils. We also found lower OC in the MAOM pool, and lower OC and N in the POM pool, though these were not significantly lower than in GSU soils. Typically, the MOAM pool is a large N store because MAOM is composed primarily of microbial products which are rich in N (Cotrufo et al., 2019). This suggests that the addition of anthropogenic materials influenced microbial activity and nutrient cycling, significantly reducing N storage in the MAOM pool, and possibly also altering the POM N and OC stores.

Anthropogenic additions to urban soils include a range of materials such as construction rubble, concrete, brick, glass, plastic and metal; though the GSA soils studied lacked the quantities of black C that was observed in SA soils. Urban soils often have high pH values, a result of additions of concrete and carbonate materials (Washbourne et al., 2015; Pouyat et al., 2020; O'Riordan et al., 2021a). Dissolved IC was significantly higher in GSA soils than GSU soils and the soil pH was also significantly higher. Decomposition and N mineralisation is determined by physico-chemical properties, litter inputs, microbial demand, pH and the climate (Risch et al., 2019). It is possible that the additions altered soil properties, including pH, making the soil less preferable for microbes, reducing decomposition and reducing replenishment of nutrients to the soil. Reduced decomposition would prevent OM losses initially, though over time, reduced release of nutrients would lead to less plant and root growth, less OM inputs to soil and lower OC and N content, as seen in our results. We also found significantly less dissolved OC in GSA soils, indicative of low available OM (Smreczak and Ukalska-Jaruga, 2021), further supporting the theory that decomposition was reduced in the greenspace soils due to the additions. It would be beneficial to investigate this further in future studies and determine whether there is an impact on primary production in greenspace soils with anthropogenic additions.

Alternative causes for the low MAOM N content could be altered leaching, denitrification or immobilisation. Studies on urban turfgrass have found lower than expected nitrate leaching and N_2O emissions from turfgrass ecosystems (Qian and Follett, 2012) and lawns can even be a sink for atmospheric N deposition (Raciti et al., 2008). However, the GSA soils had a coarser texture than undisturbed greenspace soils as a result of the additions. Coarser soils are more likely to experience mineral N leaching during rainfall (Gaines and Gaines, 1994) and this may have led to some N loss. It is unlikely that these soils experienced significant waterlogging or denitrification due to the coarser texture.

In both greenspace soils (GSU and GSA), the SOM patterns were typical of grassland soils where MAOM OC and N pools are larger than POM OC and N pools (Cotrufo et al., 2019). This shows the importance of urban greenspaces in providing stable soil OC stores that persist with long residence times, while also highlighting that anthropogenic additions to greenspace soils appear to reduce OC and N storage. It also illustrates that the additions to greenspaces were different to those in sealed soils and therefore had different outcomes for OC and N storage. In the USA alone, 1.9% of land (16 million ha) is turfgrass (Qian and Follett, 2012), illustrating the opportunity of urban greenspaces to contribute to stable long-term urban soil OC storage, and the importance of studying the impacts of anthropogenic additions on this.

4.4.4 Inorganic C in sealed and greenspace soils

Dissolved IC was particularly high in the sealed soils, likely a result of the carbonate containing materials used in roading building and sealing surfaces, such as concrete and limestone. In soils with low precipitation, carbonates and cations released by weathering can accumulate due to the lack of leaching and can cause soil to become alkaline (Zamanian et al., 2016; Kim et al., 2020). In sealed soils with limited precipitation and high levels of carbonate materials, it is likely these minerals are accumulating. Anthropogenic additions to sealed soils did not lead to higher dissolved IC content, indicating that all sealed soils were exposed to carbonate materials due to the proximity to construction materials, whether there were direct additions to soil or not. It has been suggested that dissolved CO_2 in soil water may precipitate as carbonate on biochar surfaces that have a high pH and abundant metals ions (Lehmann et al., 2011). However, the presence of black C in SA soils did not lead to greater dissolved IC, and the lack of regular water input to sealed soils suggests that this process was not occurring in SA soils.

The patterns of dissolved OC and IC were much more affected in greenspace soils than in sealed soils. In greenspace soils with additions (GSA), there was significantly more dissolved IC than in GSU soils. It is possible that this is a result of enhanced weathering in the presence of carbonate forming minerals (Renforth and Manning, 2011; Washbourne et al., 2015; Beerling et al., 2020). It is likely that additions to the GSA soils contributed Ca and Mg minerals which derive from broken down concrete, limestone and dolomite (Washbourne et al., 2012). The greenspace soils are likely to have more dissolved CO_2 due to the higher SOM content and occurrence of decomposition in these soils. Dissolved CO_2 in the soil can bind with the Ca and Mg minerals to form carbonates, effectively trapping the CO_2 as a precipitate and storing C in inorganic form (Berner and Lasaga, 1989; Renforth and Manning, 2011). The GSA soils also exhibited low dissolved OC content which suggests it may have been depleted due to carbonate precipitation, leading to the high IC content observed. While it is thought additions to greenspace soils may negatively affect decomposition and microbial activity (and OC storage) it may be beneficial for absorption of CO_2 and storage as carbonate.

4.5 Conclusion

This study suggests that there are numerous effects of urbanisation on soil C persistence in urban sealed and greenspace soils. We set out to test the hypotheses that, (i) sealed soils will have less OC in MAOM than greenspace soils and therefore less persistent OC; and (ii) soils with anthropogenic additions will have more OC in the MAOM and therefore more persistent OC in both sealed and greenspace soils. We found that anthropogenic additions had differing effects depending on whether soil was from urban greenspaces or from under sealed surfaces. In sealed soils without additions, sealing negatively affected soil OC storage across both the accessible POM pool and the more stable MAOM pool. These results illustrate the depletion of the long-term persistent MAOM OC store in sealed soils, and therefore, hypothesis i was accepted. In soils with additions, the effect of sealing was somewhat compensated for by the addition of legacy black C during road construction which led to high OC in the POM pool. Despite this, OC in the MAOM pool remained low. Therefore, hypothesis ii was rejected for sealed soils as the results illustrated depleted stores of persistent OC despite additions. This black C is vulnerable to loss by removal during construction work or decomposition. In addition, it is likely that the black C did not contribute to microbial activity in sealed soils and therefore did not contribute to the long-term microbially-derived MAOM OC store. This has implications for the use of biochar in urban soils, and future studies into its effect on OC storage in sealed soils would be beneficial. In general, further studies into the ecological processes occurring in sealed soils would build a greater understanding of OC storage and persistence and enable better long-term management of soil OC during urban construction and development.

In greenspace soils, the balance between POM and MOAM OC and N storage showed the typical pattern for grassland soils which illustrates the importance of urban greenspaces in providing persistent soil OC with long residence times. The effect of anthropogenic additions in greenspace soils did not lead to greater OC in the MAOM pool, and conversely led to lower N stock, lower N in the MAOM pool, and lower dissolved OC. Therefore, we rejected hypothesis ii for greenspace soils as additions did not lead to greater persistent OC. This is likely a result of the additions causing a reduction in decomposition, leading to lower stores of N-rich microbial OM in the MAOM pool, and lower OC across the POM and MAOM pools. This suggests that the additions could pose a threat to ecological processes such as decomposition and long-term N and OC storage in greenspace soils. Future studies should investigate this further by studying the effect on primary productivity in greenspace soils with additions. Finally, it is possible that the presence of Ca minerals in anthropogenic additions to greenspace soils leads to enhanced weathering and storage of C as inorganic carbonate, illustrating that the effect of additions to greenspace soils may lead to mixed outcomes.

5. The influence of depth and soil history on soil carbon under sealed surfaces

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Abstract

In urban soils sealed with impervious surfaces, soil carbon is widely considered to be depleted, though recent studies have suggested this is not always the case. Deep soils in urban areas have rarely been studied, and less still is known about deep soils under sealed surfaces, though subsoils are increasingly of interest for their carbon storage capacity. In addition, urban soils are highly altered by the history and development of cities, and this context needs to be taken into account when considering urban soil carbon. Despite this, there is limited knowledge on how the history of urban soils influences their carbon storage over depth. In this study, we present an investigation into three sealed soil profiles of 1 m depth in Manchester and Salford, UK. We study the distribution of carbon and nitrogen over depth and the effect of soil history. Our results illustrate that in sealed soils, the history and development of the area is a key controlling factor in determining carbon stores. We found that soil history had a greater influence over soil carbon and nitrogen stocks than depth. The pattern traditionally seen of a carbon decline over soil depth was not seen in these urban soils. Cultural layers built up through successive development histories led to consistent and higher than expected carbon and nitrogen stocks in deeper sealed soils. In soils without cultural layers, carbon stocks also stayed consistent over depth without a decline. However, in some circumstances, development led to the removal of soil and subsequent loss of carbon stocks. This study illustrates the importance of considering the soil and development history of sealed soils over depth when assessing urban soil carbon. The approach taken in construction has implications for long-term carbon storage in sealed soils and should be taken into consideration in urban construction projects.

5.1 Introduction

Urban soils are increasingly being studied for their potential to provide important ecosystem services, such as storing large amounts of soil carbon (C) (Vasenev and Kuzyakov, 2018; Pouyat et al., 2020). Soil sealing with impervious materials in urban areas, such as tarmac and paving, has been shown to significantly limit soils ability to function. In particular, sealing has been found to reduce soil C and nitrogen (N) stores compared to greenspace soils due to a removal of topsoil (Raciti et al., 2012; Yan et al., 2015; Majidzadeh et al., 2017; Lu et al., 2020), as well as negatively affecting the soil microbial community and activity (Piotrowska-Długosz and Charzyński, 2015; Pereira et al., 2021). However, some sealed soils have been shown to have higher amounts of C than previously assumed (Edmondson et al., 2012; Cambou et al., 2018; Bae and Ryu, 2020; O'Riordan et al., 2021a - chapter 3).

Studies of soil C have mostly focused on topsoil as this is where C content is usually highest. However, there is increasing interest in the importance of deeper soils for C storage as considerable amounts of C can be stored across the whole soil profile, and depth plays an important role in determining C stores (Jobbágy and Jackson, 2000; Salomé et al., 2010; Rumpel and Kögel-Knabner, 2011; Li et al., 2016; Herrmann et al., 2018; Simo et al., 2019). In urban settings, there is limited knowledge on soil C in deeper soils, though they have been identified as playing an important role in urban soil C storage (Lorenz and Lal, 2005; Mazurek et al., 2016; Scharenbroch et al., 2017; Vasenev and Kuzyakov, 2018; O'Riordan et al., 2021b - chapter 2). Urban soils are highly influenced by human activities and often experience the addition of artefacts and anthropogenic materials over time as cities develop, leading to the formation of Technosols (Rossiter, 2007; FAO, 2015). Technosols have very variable properties and C content, though they have been shown to exhibit some of the highest C stocks when compared to WRB soil groups as a result of the artefacts found in the soil (Allory et al., 2022). The process of settlement history adding materials into soil also creates what are known as cultural layers (Alexandrovskaya and Alexandrovskiy, 2000; Vasenev et al., 2013; Vasenev and Kuzyakov, 2018; Bae and Ryu, 2020). These cultural layers are known to lead to both high and spatially heterogeneous soil C measurements in urban areas (Vasenev et al., 2013; Mazurek et al., 2016). In addition, soil N in deeper urban soils has received little attention. Soil N is important for C storage as a result of N limitation controlling microbial decomposition of soil organic matter (SOM) (Groenigen et al., 2006; Averill and Waring, 2018; Rocci et al., 2021), and therefore it is also necessary to consider soil N in both topsoil and deeper urban soils to understand urban soil C storage.

While our knowledge on urban deep soil C storage is limited, less still is known about C storage in sealed soils. Sealed subsoils have been shown to exhibit a different vertical SOC distribution to adjacent greenspace soils (Yan et al., 2015) and have been suggested as a significant urban soil C store (Edmondson et al., 2012; Cambou et al., 2018; Vasenev and Kuzyakov, 2018; Bae and Ryu, 2020; O'Riordan et al., 2021a). Sealed soils have a complex formation and development history and are very heterogenous in character. They are highly altered by human activities such as topsoil removal, horizon mixing, and imports of non-native soil and other materials (Herrmann et al., 2017; Herrmann et al., 2018). They can also exhibit cultural layers due to historical anthropogenic additions which can influence sealed soil C content and lead to legacy C stores (Bae and Ryu, 2020; O'Riordan et al., 2021a). Sealed soils can be dug open and re-developed during road building and utility pipe repairs, while some remain untouched for decades or even centuries. This varied development history and soil disturbance has implications for the soil oxygen, moisture and decomposition status, and this can lead to the disruption of soil C stores under sealed surfaces. Therefore, it is important to consider the development and soil history when studying urban and sealed soils (Ziter and Turner, 2018; Herrmann et al., 2020).

In this study, we investigate the effects of soil depth and soil history on sealed soil properties, including soil C and N stocks, pH, and extractable organic C (OC) and inorganic C (IC). We ask the following research questions: 1) what is the depth distribution of C and N down sealed soil profiles; and 2) what effect does soil history have on soil properties and C storage? We hypothesise that soil C and N will decrease with soil depth. We test this hypothesis by studying three 1 m deep soil profiles in the centre of Manchester and Salford, UK, under sealed surfaces. We also explore the influence of soil history and development on soil C and N storage and the overall storage capabilities for a 1 m depth sealed soil profile. We study profiles with long and varied development histories so as to understand the influence of soil history on soil C.

This research contributes to the growing body of knowledge on sealed soils and deeper subsoils, and in particular, the influence of soil history on soil C storage. It has implications for construction activities and urban re-development and their effects on C stores in sealed soils.

5.2 Methods

5.2.1 Profile sampling

Sites were identified with archaeologists at Salford Archaeology, and profile locations were chosen where there were 1 m deep soil profiles exposed due to archaeological excavations. The archaeologists also provided information on the history and development of the sites, the likely dates of soil sealing, and the artefacts identified in the soil to provide an age for profiles and the horizons.

Profiles were identified in inner city locations in Manchester and Salford, UK. Profile 1 was located at the Globe and Simpson site, part of the St John's redevelopment at the former Granada Studios site in central Manchester (figure 5.1). The profile was on the site of a former Art Deco building built in the 1930s. Prior to this, the location was a timber yard where the ground covering was unknown, but may have been a pervious material such as gravel or organic material such as straw. Profile 1 was sampled in November 2018. Profiles 2 and 3 were from the Valette Square development in Salford, to the east of Adelphi Street (figure 5.1). These profiles were on the site of a former house built in 1824. Profile 2 was underneath the courtyard at the back of the house and was sealed under stone flagstones. It was likely that the courtyard was re-developed in the early 1900s to install plumbing, indicated by the style of drainage pipes running through the soil pit, which likely led to this profile being most recently sealed in the early 1900s. Profile 3 was located underneath the main house and was sealed with brick flooring, suggesting it had been sealed and undisturbed since 1824. Both profiles 2 and 3 were sampled in June 2019.

Samples were collected from open soil pits or exposed soil faces of 1m depth. Profiles were sampled every 10 cm down to 100 cm, providing 10 depth intervals per profile. At every depth interval, a small sample was collected and a bulk density core was taken. Profiles were replicated at a distance of 1 metre apart; for profile 1, three replicate profiles were sampled (profiles 1a-1c); for profiles 2 and 3, two replicate profiles were sampled (2a-2b, 3a-3b) which were a result of constraints to site access preventing a third replicate.

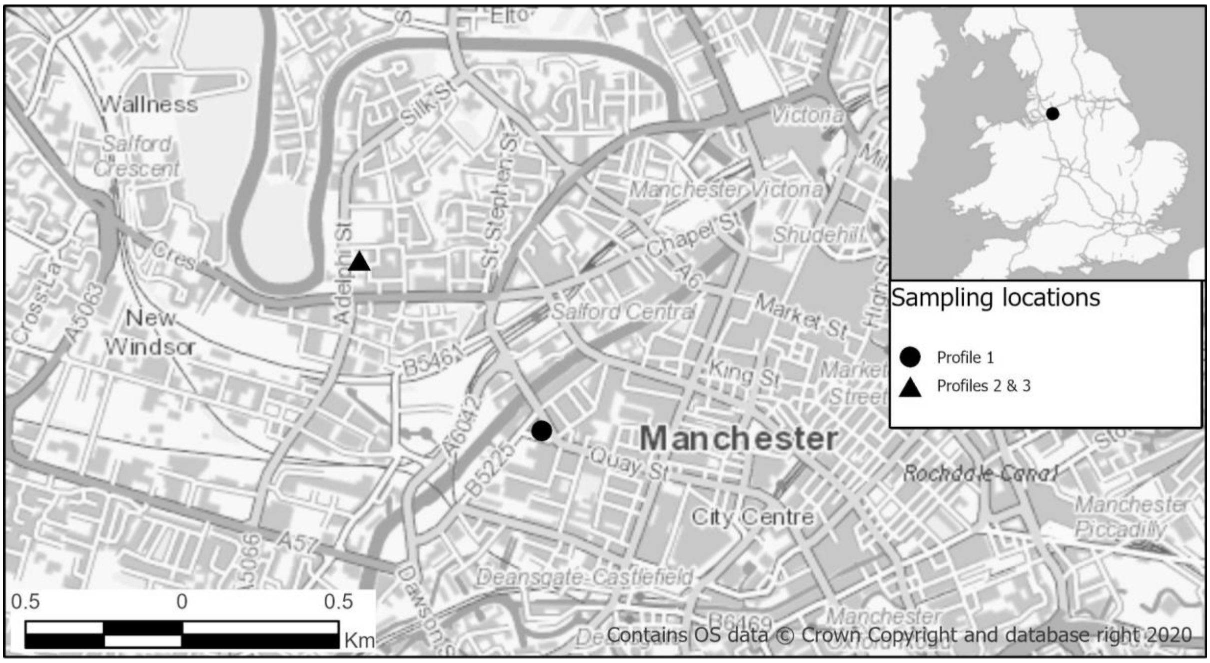
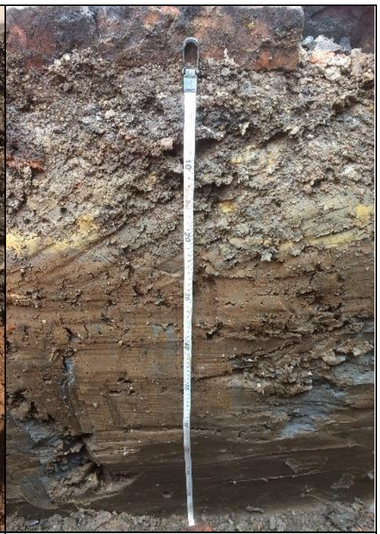


Figure 5.1: Map showing locations of profile 1 (Manchester) and profiles 2 and 3 (Salford).

5.2.2 *Profile Information*

Details about each profile and the soil history and horizons observed are set out in Table 5.1.

Table 5.1: Profile information

		
Profile 1	**Profile 2**	**Profile 3**
Profile overview		
Visible cultural layers through the depth of the profile due to successions of past use, human alteration and additions to the soil.	Cultural layers were visible in the upper profile though horizon mixing was apparent. In the lower profile, imported sand dominated.	The profile was less disturbed and cultural layers were not visible, other than some artefacts in the top 20 cm. Soil was a clay rich subsoil.
Duration of sealing		
Sealed 1930s – sealed for approximately 90 years	Sealed 1900s – sealed for approximately 120 years	Sealed 1824 – sealed for approximately 190 years
Soil history and description		
• Underneath an Art Deco building built in the 1930s. • Prior to the 1930s it was a timber yard – successive layers and compaction may have	• Underneath a courtyard sealed with stone flagstones. • Courtyard attached to a house built in 1824. • Courtyard built in 1824 but re-developed	• Underneath a house built in 1824, sealed with brick flooring. • House demolished in approx. 2000s, ground level sealed over and used as a car park.

occurred between 1800 -1930s. • Prior to this, soil was likely cultivated in a garden or for food growing. • Horizons show cultural layers through the profile. • Possible organic matter inputs from wood storage while used as a timber yard.	in 1900s, likely to install plumbing in Victorian era. • House demolished in approx. 2000s, courtyard sealed over and used as a car park. • Upper soils contained artefacts - 20th C pottery, and 19th century clay pipe and thin glass fragments, and clumps of both clay rich and more coarse soils. • Deeper soils are sand or areas of soil mixed with sand, likely imported. • Undecomposed plant roots visible between 0-70 cm.	• Artefacts found within upper layers (0-20 cm) suggesting waste materials used to level the soil before laying brick flooring. • Deeper soil appears undisturbed by human activities and is clay rich.
Soil horizons		
• 0-40 cm – soil containing artefacts (bricks, coal char, ceramics), distinctive layer of coal char at 35 cm. • 40-60 cm – clay rich soil • 60-80 cm – lighter colour, likely 17th century garden soil • 80-100 cm – dense clay rich subsoil	• 0-60 cm – soil containing artefacts (bricks, ceramics, coal char, glass) • 60-100 cm – sand dominated soil, likely imported	• 0-20 cm – soil containing artefacts (coal char, ash, brick). • 20-100 cm – dense clay rich undisturbed subsoil.

5.2.3 *Soil analyses*

All samples were passed through a 2 mm sieve and homogenised. For pH analysis, 10 g of fresh soil was mixed with 25 ml distilled water and shaken for 30 minutes at 180 rpm on an orbital shaker. It was left to settle for 30 minutes, and the pH was measured at the soil-water interface using a pH probe (Mettler Toledo, SevenCompact S220). The remaining soil was dried at 70 °C for 48 hours. Separately, the bulk density cores were weighed and dried for 48 hours at 105 °C. They were then re-weighed to determine the dry weight which was used for bulk density calculations.

To measure extractable organic C (OC) and inorganic C (IC), the 70 °C dried samples were mixed with K_2SO_4, as described by Vance et al. (1987). For this, the pH of the K_2SO_4 was checked and adjusted to between 6.8-7 using NaOH. Subsamples of 5 g dry soil were mixed with 20 ml 0.5 M K_2SO_4, and this was shaken on an orbital shaker for 1 hour at 180 rpm. This was filtered through a Whatman no. 42 filter and filtrate was diluted 1 part to 8 parts Milli-Q water. This was then analysed for extracted total C and inorganic C, using a Total Organic Carbon (TOC) analyser (Shimadzu TOC-L CPN TN). Extractable OC is calculated as the extractable total C minus the extractable IC.

For analysis of total C and N, subsamples were re-dried at 105 °C for 24 hours and then ball-milled. Subsamples were then analysed for total C and N concentration using the dry combustion method using a CN analyser (El Vario analyser, Elementar, Hanau, Germany). The C:N ratio is determined by the C/N content. Total C and N stocks were calculated using the total C and N concentrations and the bulk density values for a depth of 10 cm, using the method given by the (FAO, 2018). For profile 3, bulk density values were only available for replicate 3a and so these were used for calculations of C and N stock for both replicates 3a and 3b.

5.2.4 *Data analysis*

The three profiles enable a comparison of characteristics over depth and the influence of soil history. While each profile had been sealed for a different length of time, we did not consider the duration of sealing as a controlling factor because the influence of soil history appeared to dominate the profile characteristics. Therefore, we firstly consider the pattern of C and N over depth for each profile, then secondly consider the differences between the profiles in relation to their history and the impact of that on C and N stocks.

For the study of depth, replicates of each profile are presented individually as profiles 2 and 3 had only 2 replicates each, preventing the use of the mean. Presenting the results in this way allowed consideration of the variation between replicates and served as a useful observation of the heterogeneity of sealed soils. For the study of soil history, values were summed to get an overall value across 0-100 cm for each profile replicate. This was undertaken for the variables of C and N stock and extractable OC and IC content. For pH and C:N ratio, the median and upper and lower quartiles for each replicate are presented in boxplots.

5.3 Results

5.3.1 Depth distribution across profiles

The three profiles exhibited different patterns of C and N across depth, with only profile 2 exhibiting a clear decline of C and N stock over the 1 m profile (figure 5.2). In profile 1, the C and N content showed high variability between replicate profiles, though each replicate followed very similar C and N patterns over depth, including in extreme values. Within replicates, there was a lot of variability in values between 0-40 cm, with peaks in C and N also occurring at 60 and 70 cm. Other than these extreme values, C and N content stayed fairly consistent until 80 cm where they declined (figure 5.2 a, c). The C and N stocks followed very closely the pattern of C and N content within each profile and also declined at 80 cm (figure 5.2 b, d). The profile had a mean C stock of 2.54 ± 1.79 kg m^{-2} per 10 cm depth interval; while the mean N stock was 136.83 ± 46.56 g N m^{-2} per depth interval. The C:N ratio declined slightly over depth, illustrating that there was more N in relation to C lower down the profile (figure 5.3 c). Extractable OC content in profile 1 also followed a very similar pattern to C and N for each replicate, with peaks identified at 40 cm, 60 cm and 70 cm that correlate with high values in C and N content, and a similar decline at 80 cm. In contrast, none of the replicates of profile 1 exhibited any extractable IC other than the top 10 cm of profile 1c (figure 5.3 b). The pH of profile 1 was acidic with values consistently below 6 for replicates 1a and 1b, and often below 6 for 1c.

In profile 2, the C and N content showed some variability between 0-60 cm and a peak at 40 cm in replicate 2b, though below 60 cm both C and N content exhibited a clear decline which persisted until 100 cm (figure 5.2 a, c). The C and N stocks followed a similar pattern with the same variability between 0-60 cm and a marked drop off at 60 cm, exhibited clearly by both profile replicates. This led to a mean C stock of 1.74 ± 1.33 kg C m^{-2}, and a mean N stock of 57.21 ± 41.49 g N m^{-2} per 10 cm depth interval. The C:N ratio slightly decreased over the profile depth, though there was variation between the two replicates. Despite this, both exhibited an increase in C:N ratio at 80 cm (figure 5.3 c). Extractable OC followed the same pattern as C and N stocks and also exhibited a notable decrease at 60 cm (figure 5.3 a). Extractable IC in profile 2 followed a similar pattern to the C and N stocks with similar variability through the upper profile. However, lower down the profile at 80 cm the replicates differed, with 2a decreasing and 2b increasing in IC. The pH of profile 2 did not appear to be altered by depth and was quite alkaline through the whole profile, staying consistently between pH 8 and 9 across depth (figure 5.3 d).

In profile 3, the C and N content were highest in the top 20 cm of the profile and then decreased to a consistent level between 20-100 cm. The C and N stocks also followed this pattern, exhibiting relatively stable stocks across the profile after a decrease below 20 cm, though the C stock was more varied across depth in profile 3b (figure 5.2 b, d). The mean C stock was 2.75 ± 0.75 kg C m^{-2}, and the mean N stock was 113.19 ± 29.79 g N m^{-2} per 10 cm depth interval. Extractable OC followed a very similar pattern to C and N content, reducing below 20 cm and staying at a consistent level across the remaining profile (figure 5.3 a). In contrast, the extractable IC initially decreased after 10 cm, then increased at 30 cm and remained at a relatively high level through the remaining profile (figure 5.3 b). The pH in profile 3 remained consistently alkaline, staying between 8 and 9 across depth other than a brief drop below pH 8 at 20-30 cm (figure 5.3 d).

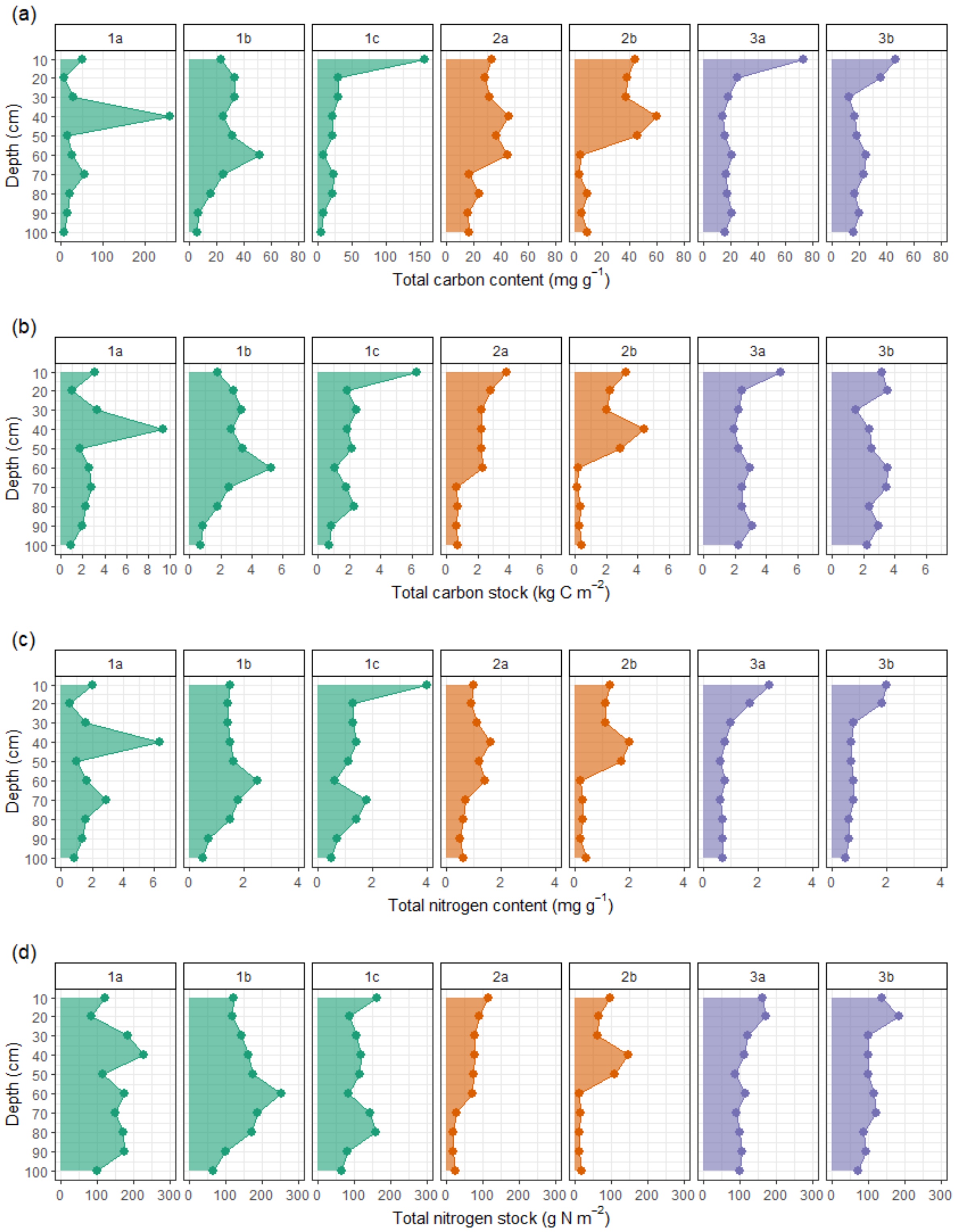


Figure 5.2: Depth plots showing (a) total carbon content (mg g^{-1}); (b) total carbon stock (kg m^{-2}); (c) total nitrogen content (mg g^{-1}); and (d) total nitrogen stock (g m^{-2}) across profile depth. Profiles shown are profiles 1a-1c, 2a-2b and 3a-3b. Note the different scale used for profiles 1a and 1c to allow for extreme values.

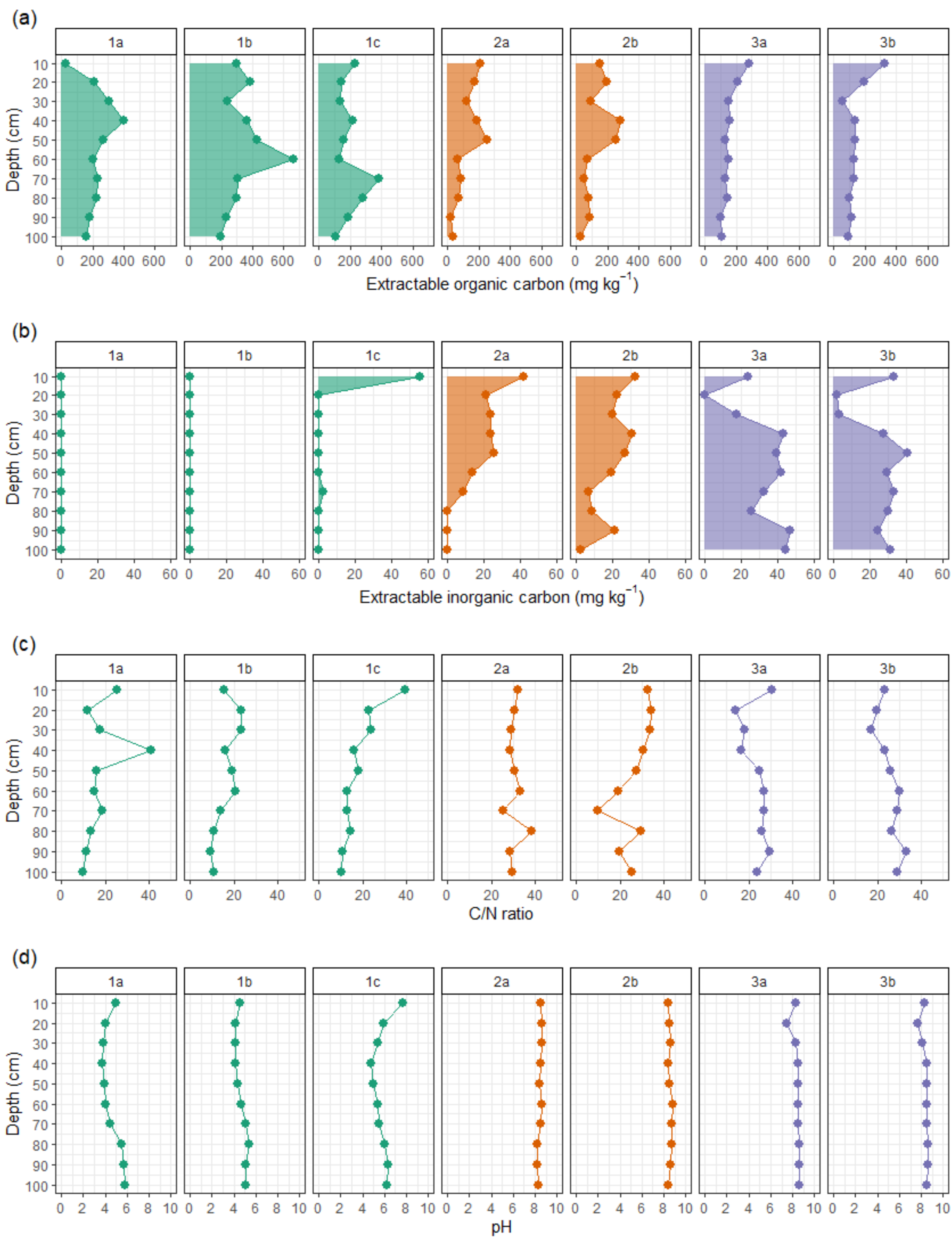


Figure 5.3: Depth plots showing (a) extractable organic carbon (mg kg^{-1}); (b) extractable inorganic carbon (mg kg^{-1}); (c) C:N ratio; and (d) pH across profile depth. Profiles shown are profiles 1a-1c, 2a-2b and 3a-3b.

5.3.2 *The influence of soil history*

Over the whole profile depth (0-100 cm) profiles 1 and 3 had similar C stocks, with profile 1 exhibiting 25 ± 4 kg C m^{-2} overall, and profile 3 replicates ranging between 27 and 28 kg C m^{-2}. Profile 2 had the lowest overall C stock, with replicates ranging between 16 and 18 kg C m^{-2} (figure 5.4 a). In profile 3 approximately half (49-53%) of the overall C stock in 0-100 cm was found below 50 cm depth. In profile 1, 38% of the overall C stock was below 50 cm, while in profile 2, between 8-27% was below 50 cm.

N stocks followed a similar pattern to C stock though profile 1 exhibited greater N than profile 3, with 1,368 ± 219 g N m^{-2} overall in profile 1, and replicates ranging between 1,098 and 1,166 g N m^{-2} in profile 3 (figure 5.4 b). Profile 2 exhibited the lowest N stock overall, with replicates ranging between 545 and 599 g N m^{-2}. This was reflected in the C:N ratio which showed that profile 1 had consistently lower C:N ratios than profiles 2 and 3, indicative of the higher N content in profile 1 (figure 5.4 c).

Across the whole profile, extractable OC was greatest in profile 1, though there was some variability across the profile replicates. Despite this, profile 1 had at least twice the amount of extractable OC as the other profiles, with a mean of 2,554 ± 791 mg kg^{-1} OC over 0-100 cm, compared to profile 2 (between 1,067 and 1,102 mg kg^{-1}), and profile 3 (1,157 and 1,251 mg kg^{-1}) (figure 5.4 d). Though there was variability in extractable IC content across the profile replicates, the greatest IC was found in profile 3, while profile 1 exhibited very little, with 2 of the 3 replicates having none (figure 5.4 e). This is reflected in the notably lower pH values across profile 1, while profiles 2 and 3 had similar higher pH values of between 8 and 9 (figure 5.4 f).

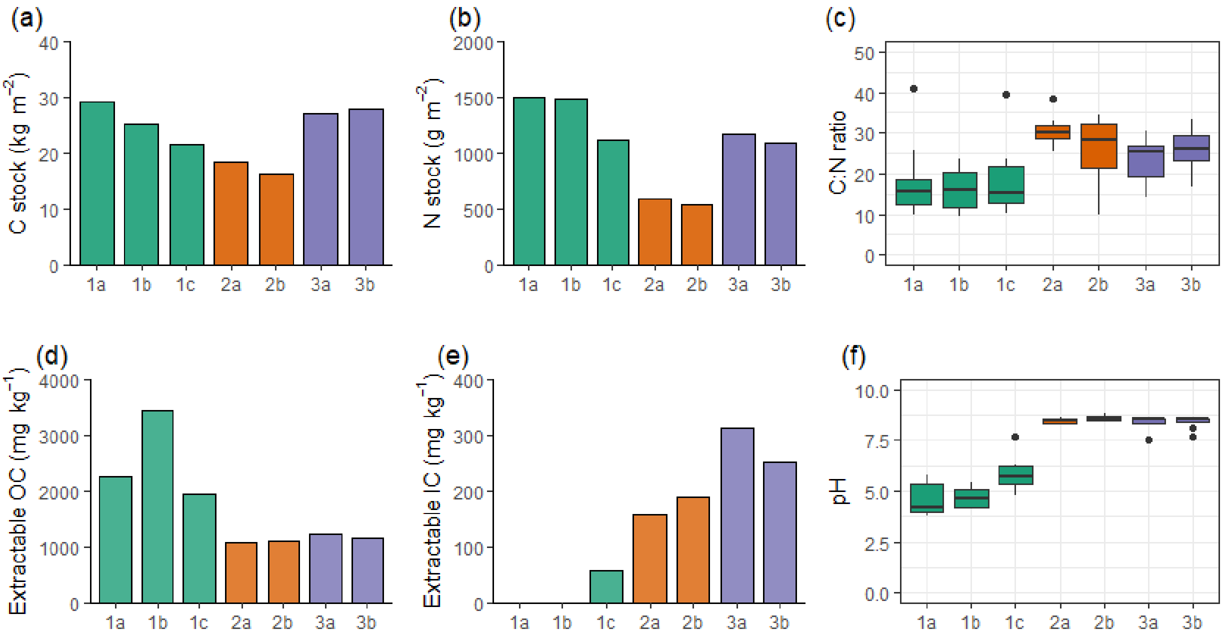


Figure 5.4: Figures a, b, d and e show the sum of C and N across the whole profile (0-100 cm), showing (a) carbon stock (kg m-2); (b) nitrogen stock (g m^{-2}); (d) extractable organic carbon (mg kg^{-1}); and (e) extractable inorganic carbon (mg kg^{-1}). Boxplots show (c) C:N ratio; and (f) pH value, where the box represents the upper and lower quartiles, whiskers show upper and lower most values, and horizontal lines show the median. Circled data points show outliers at 1.5 times the interquartile range. Profiles shown are profiles 1a-1c, 2a-2b and 3a-3b.

5.4 Discussion

In this study we hypothesised that soil C and N would decrease with soil depth. The results illustrate that C and N did not decrease over depth as is typically seen in non-sealed soil profiles (section 5.4.1), and therefore, the hypothesis was rejected. We also explored the influence of soil history and development on C and N stocks, and how the soil histories, former land uses and soil treatments could result in cultural layers and losses of soil, with varying impacts on C and N storage (sections 5.4.2, 5.4.3, 5.4.4).

5.4.1 Soil history controls C and N storage rather than depth

We found that C and N stocks did not necessarily decrease with depth in these sealed soil profiles. In profiles 1 and 3, C and N stocks were fairly consistent throughout most of the profile depth, with profile 1 exhibiting some very high values across depth. Profile 2 displayed a decline in C and N stocks after 60 cm which correlated with a removal of the soil and replacement with sand dominated material. These observations are in contrast with most studies on deeper soils which typically show a decrease in soil C and N stocks with increasing depth (Fang and Moncrieff, 2005; Lorenz and Lal, 2005; Rumpel and Kögel-Knabner, 2011; Bai et al., 2016; Li et al., 2016). The decrease in C and N that is typically seen in non-sealed soils is a result of lower C content in subsoils in comparison to topsoils, where the organic matter and C and N concentrations are much higher (Rumpel and Kögel-Knabner, 2011). In this study, the profiles are located in the inner city centres of Manchester and Salford and have been influenced by human development for hundreds of years. These sealed soils have been so altered by development history that this appears to be the key controlling factor determining C and N stocks within the soil profile rather than depth. This is a pattern that has also been observed in other studies of urban soils, where C stock did not decline over depth due to the result of human alterations and additions to the soil (Scharenbroch et al., 2017; Bae and Ryu, 2020; Allory et al., 2022).

5.4.2 Cultural layers can lead to high C and N stores

Profiles 1 and 2 have been altered by successions of development that have added materials and artefacts into the soil and have led to the formation of cultural layers. In profile 1, cultural layers were visible across the whole profile down to 80 cm depth. The C stocks between 40-100 cm were 12 ± 3 kg C m^{-2}, nearly twice that observed in sealed soils at the same depth in Leicester, UK, where soils beneath roads had 6.7 kg C m^{-2} (Edmondson et al., 2012). In profile 2, artefacts between 0-60 cm helped to identify the cultural layers as a mixture of 19^{th} and 20^{th} century in age. In these layers there were both high and variable C and N contents, and overall C stocks for the 0-60 cm layers were between 15 and 16 kg C m^{-2}. It is likely that the artefacts and cultural layers observed contributed to the C stocks in these profiles as it known that these features lead to increases in soil C stores as well as high variability (Vasenev et al., 2013; Vasenev and Kuzyakov, 2018; Bae and Ryu, 2020; Allory et al., 2022).

In profile 1, artefacts identified between 0-30 cm included rubble, brick, coal char, and ceramics. Below this was a distinctive layer of coal char at 40 cm which led to very high C content at this depth (figure 5.2 a). Profile 2 also exhibited coal char distributed through the cultural layers between 0-60 cm. Coal char, or black C, results from the incomplete combustion of fossil fuels and is common in urban soil, sometimes accounting for up to 70% of the SOC in urban soils (Lorenz et al., 2006; Edmondson et al., 2015; Schifman et al., 2018). Along with other rubble and brick waste materials, coal char has often been used to level soils prior to constructing roads and buildings or as a fill material (MacBride, 2013; Mazurek et al., 2016), and therefore, likely contributed to the soil C stocks in these cultural layers.

Older cultural soils were observed lower down the profile in profile 1. Between 40-60 cm there was a layer of clay rich soil, and below that at 60-80 cm, there was likely former garden or cultivated soil, both of which had similar C and N stocks to those higher up the profile. Although artefacts were less visible in these layers, the soil still maintained consistent C and N stocks. This is typical of urban cultural layers which exhibit fewer artefacts lower down the profile, though C can remain higher due to black C content which has moved through the layers and is enriched at depth (Glaser et al., 2000; Mazurek et al., 2016). In profile 1, however, black C would not explain the high N and extractable OC contents, which indicate that the C and N stocks relate to other historical additions or ecological contributions of C. Prior to sealing in the 1930s, the site for profile 1 was a timber yard which may have led to organic matter being added into the soil. Wood from construction is a common artefact found in cultural layers (Alexandrovskaya and Alexandrovskiy, 2000; Lorenz and Lal, 2005; Mažeika et al., 2009; Alexandrovskiy et al., 2012) and may have led to higher N and extractable OC contents.

The N contents in profile 1 were particularly high for a sealed soil, exhibiting a mean of 1.62 ± 1.15 mg g^{-1} per depth interval. This was notably larger than previous studies of urban soil in Manchester, where mean N content was 0.62 ± 0.23 mg g^{-1} in sealed soil, and 1.41 ± 0.50 mg g^{-1} in greenspace soil (O'Riordan et al., 2021a). Extractable OC was also fairly high for a sealed soil in profile 1, exhibiting a mean of 255.42 ± 121.32 mg kg^{-1} per depth interval. This is higher than in previous studies where sealed soil exhibited 119.98 ± 67.81 mg kg^{-1} in Manchester, UK (O'Riordan et al., 2021a), and less than 80 mg kg^{-1} in Alabama, USA, in soil sealed for 114 years (Wang et al., 2021). In contrast to topsoil DOM, which is mostly plant-derived, subsoil DOM is largely microbial in origin (Guggenberger and Zech, 1994; Kaiser et al., 2004), and is often dominated by N-rich compounds (Kaiser and Kalbitz, 2012). As microbial products tend to be higher in N (Cotrufo et al., 2019), the higher extractable OC and N contents suggest there has been, and may still be, microbial activity in this profile, likely enabled by nutrients deposited in the cultural layers. Though microbial activity tends to decrease with depth, subsoils do remain metabolically active and can contain substantial numbers of microorganisms (Taylor et al., 2002; Rumpel and Kögel-Knabner, 2011). In addition, profile 1 was found to be acidic, with pH levels generally below pH 6. Dissolved CO_2 from decomposition of organic matter forms a weak acid (Weil and Brady, 2017) which supports the prospect of decomposition having occurred in this profile. This would also explain lack of extractable IC found in profile 1, as carbonates become dissolved in acidic conditions (Guo et al., 2016).

In profile 2, the high C and N stocks seen in the cultural layers are not accompanied by the high extractable OC content as seen in profile 1. The higher C:N ratio and alkaline soil conditions found in profile 2 also suggest that the same C and N dynamics and soil processes may not be occurring as suggested in profile 1. There were also undecomposed plant roots visible in the cultural layers indicating that plants growing in the courtyard at the profile site may have contributed to C stocks through their roots, though their presence suggests that decomposition has not been occurring recently in profile 2. In addition, profile 2 exhibited extractable IC that followed the general pattern of the C and N stocks, a trend not seen in either profile 1 or 3. This highlights the heterogeneity found in urban soils, and in particular the variation brought about by cultural layers and artefacts. It is possible that this profile contained more Ca^{+} bearing materials, such as plaster from construction rubble, concrete or limestone, all of which can lead to high CaCO3 in cultural layers (Mazurek et al., 2016; Vasenev and Kuzyakov, 2018).

5.4.3 Soil removal causes C and N losses

Below the cultural layers in profile 2, the C and N stocks notably decreased below 60 cm where the soil appeared to have been amended or replaced with sandy material. The profile sits underneath what was a courtyard behind a house, and the removal of soil may have been during redevelopment of the courtyard or installation of plumbing for the house in the early 1900s. It is likely this redevelopment led to the loss of native soil below 60 cm, and the re-use of some of the excavated soil, or other soil, above 60 cm which led to the horizon mixing that can be seen in the cultural layers (Table 5.1).

In this profile, the replacement of soil with sand led to the lowest overall C stocks of all the profiles, with replicates ranging from 16 to 18 kg C m^{-2} over 0-100 cm. Sandy soils tend to have lower C, while silt and clay soils are able to retain more C by binding with soil minerals (Tiessen and Stewart, 1983; Yost and Hartemink, 2019). Therefore, this replacement illustrates the impact of the removal of soils, and particularly cultural layers, in urban areas. The removal of soil is an issue that continues in contemporary construction and often leads to large quantities of soil being sent to landfill (Green Construction Board, 2020). As cultural layers are known to contribute high C stores, the loss of these urban soils compounds the loss of soil C due to redevelopment.

The widespread loss of urban soil horizons, in particular intermediate B horizons between 50-100 cm, has been observed across multiple cities in the USA and has been linked to soil removal, horizon mixing, and imported fill additions (Herrmann et al., 2018). These intermediate horizons are usually accumulation zones from the A layer above and are important for soil functions such as adsorbing nutrients and dissolved OM (Kalbitz et al., 2000), water regulation (Caldwell et al., 1998) and C storage (Salomé et al., 2010). While the study by Herrmann et al. (2018) considers both greenspace and fill soils on brownfield sites, the loss of soil may be even more common in sealed soils and indicate a wider problem for urban soil functioning. Future research should consider the impacts of soil removal and loss due to development, and the wider impacts of this on urban soil functions such as carbon storage. The loss of soil C stores due to construction is not widely considered by either the research or practice communities, and therefore, inclusion of this in planning requirements such as environmental impact assessment or C budgeting would be beneficial.

5.4.4 *Sealed subsoils are not necessarily depleted of C*

Profile 3 exhibited little disturbance and had been sealed for the longest duration of time, since approximately 1824. While it had some artefacts and coal char in the top 20 cm, it did not display obvious cultural layers below this. It may be expected that this soil would have low soil C and N stocks due to the long duration of sealing, however it exhibited low, yet consistent, C and N stocks and as a result had similar stocks to profile 1. The overall C stocks (0-100 cm) ranged from 27 to 28 kg C m^{-2}, which were only slightly less than the mean C stocks in greenspaces in the Chicago region at equivalent depths, at 36.4 ± 1.4 kg C m^{-2} (Scharenbroch et al., 2017). While subsoils under sealed surfaces have rarely been considered, one recent study found that deep sealed soils can have high C stocks which can increase below 1 m, with C stocks being between 7 and 14 kg C m^{-2} at 1-2 m depth (Bae and Ryu, 2020). We also expected the C stocks to decline over depth as this is the commonly observed pattern over depth (Lorenz and Lal, 2005). However, as with profiles 1 and 2, we did not observe this pattern in profile 3, despite its lack of visible cultural layers.

Profile 3 displayed properties that are characteristic of a subsoil, with high clay content and bulk density and high extractable IC. Clay soils typically store most C as mineral-associated OM which has long residence times and is protected from microbial decomposition in organo-mineral complexes (Paul, 1984; Hassink, 1997; Six et al., 2002). Subsoils also typically have greater IC which contributes to total C stock, and is a result of the presence of Ca^{+} ions and a high pH environment, also seen in this soil. Carbonates can accumulate down the soil profile as Ca^{+} is carried in soil water and precipitates as $CaCO_3$ deeper in the soil profile (Meyer et al., 2014). It is also known that black C migrates down the soil profile and can accumulate in deeper soils (Glaser et al., 2000). Black C is common in urban soils and has long residence times (Rumpel and Kögel-Knabner, 2011; Edmondson et al., 2015), and as this profile had coal char additions at 0-20 cm, it is possible some of this had migrated into lower layers and contributed to the total C stock.

Dissolved OM (DOM) is also a key source of C in subsoils (Rumpel and Kögel-Knabner, 2011; Kaiser and Kalbitz, 2012). It has been shown that DOM in sealed soil can be depleted of various biochemical groups (Wang et al., 2021). As sealed soils receive limited OM from plant roots or root exudates, it is likely that DOM in sealed soils will be derived from extant OM or microbial products. DOM usually migrates downward through soil and becomes either mineralised or stabilised with clay minerals, while only a small amount is leached out (Kaiser and Kalbitz, 2012). DOM that becomes stabilised as mineral-associated OM could have mean residence times of 100–200 years (Tipping et al., 2012). Although we observed low extractable OC content in profile 3, it is possible that past decomposition may have provided DOM which has since been sorbed onto clay minerals and forms the current C stock. It is also possible that the lack of fresh OM inputs to the soil have led to a relatively dormant decomposition state, given that accessible substrate is low and microbial activity is probably low. Microbial activity in subsoils is known to decrease with depth (Taylor et al., 2002; Fang and Moncrieff, 2005), and also under sealed surfaces (Zhao et al., 2012; Wei et al., 2014b; Piotrowska-Długosz and Charzyński, 2015; Pereira et al., 2021), and therefore this would lead to a lack of decomposition of the mineral-protected OC.

Profile 3 illustrates that, over depth, sealed soils can be a large and consistent store of soil C, even where soil and development history has not contributed large anthropogenic C stores. This needs to be taken into account when considering both soil C budgets and soil mapping, and during urban redevelopment involving the storage or removal of soil.

5.5 Conclusion

This study set out to explore the effect of depth and soil history on soil C and N storage. We tested the hypothesis that soil C and N will decrease with soil depth. We found that soil C and N stocks do not always decrease over depth in sealed soils and that soil history appears to have a greater influence over soil C and N than depth. Therefore, we rejected our hypothesis. Sealed soils were so altered by historical developments, whether that was the addition of artefacts and creation of cultural layers, mixing of horizons, or the removal of soil during construction, that they seemed to determine the C and N status of the soil. In contrast to natural systems where the pattern of soil C and N may be more predictable over depth, we cannot have the same expectations for historically influenced sealed soils in urban areas. The long soil histories and addition of artefacts to the profiles led to the creation of cultural layers which provided particularly high C stocks across depth. In sealed soils without cultural layers there were still consistent C stocks which, over depth, resulted in notable C stores. These were attributed to the clay-rich and highly dense subsoils which provided deep stores of soil C. Conversely, soil C losses were observed due to the removal and replacement of soil with imported materials during redevelopment. This highlights the impact of construction activities and the loss of soil C stores during development which remains a common and large-scale problem in contemporary construction practices.

We have illustrated that in deep sealed soils, with or without cultural layers, C stocks remain notable and are not depleted, and soils with cultural layers may have higher soil C. Therefore, it is important that sealed soil C is taken into account in urban soil C budgeting and soil mapping. It is also necessary to consider the soil C cost of construction due to the removal of deep historical soils, and the prevention of this should be sought through adequate inclusion of soil C in environmental impact assessment and project C budgets.

5.6 Acknowledgements

We would like to extend our thanks to Salford Archaeology, and in particular to Graham Mottishead for his help in identifying sites, providing access for sampling and, not least, for the wealth of information and insight he provided into the age and history of profiles and the artefacts found within the soils.

6. General Discussion

6.1 Summary of aims, objectives and hypotheses

The growing interest in urban soil to provide ecosystem services and, in particular, to store C has led to increased studies into urban soil C. However, urban soil research is a young field and there remained large gaps in our understanding of urban soil functioning, ecosystem services, and C storage. In particular, there was limited knowledge on soil C storage in sealed soils, on the stability and persistence of that C following sealing, and how is it distributed over depth.

This thesis sought to investigate the effect of urbanisation and soil sealing on soil ecosystem services and soil carbon. The thesis aimed to (1) improve our understanding of urban soil's role in providing ecosystem services; and (2) investigate the effects of soil sealing on the ecosystem services of soil C and nutrient storage. To meet these aims, the following objectives were addressed:

1. The literature on ESs provided by urban soils was reviewed to build a picture of the current knowledge base and identify research gaps (chapter 2);
2. The effect of soil sealing on soil C and nutrient storage was investigated by developing a dataset across sealed and greenspace urban soils (chapter 3);
3. The effect of sealing and anthropogenic additions on soil C persistence was determined using analysis of functional pools of soil organic matter (chapter 4);
4. The influence of profile depth and soil history on the distribution of C and N was investigated in sealed soils with varied development histories (chapter 5).

To meet these objectives, the thesis consisted of one review chapter and three data chapters as follows:

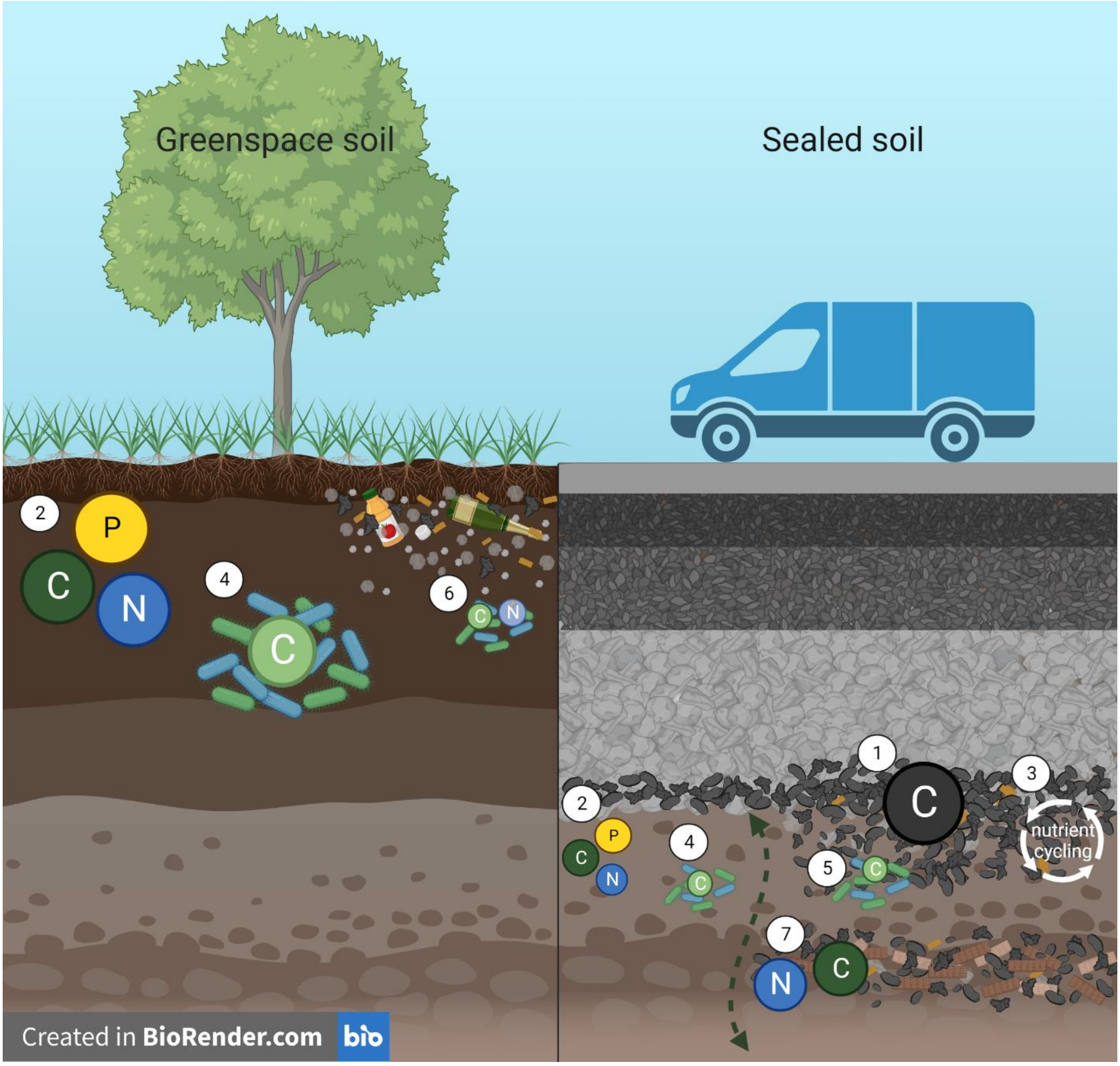
Chapter 2 addressed **objective 1** and provided a systematic review of the literature on urban soil ESs to create a broad picture of current research. It found that supporting and regulating services including soil biological activity, nutrient cycling and C storage were most studied. However, cultural, food and water-related services were less studied. It identified urban soil multifunctionality as a key direction for future research, along with more global studies, community integration and potential future drivers of change.

Chapter 3 addressed **objective 2** by investigating the effect of sealing on soil C and nutrients. It tested the hypotheses that sealed soils have (i) lower soil C stocks, (ii) lower soil nutrient stocks, and (iii) altered nutrient dynamics compared to greenspace soils. Both hypotheses i and iii were rejected, as where soils had anthropogenic additions they did not exhibit lower C stocks or altered nutrient dynamics compared to greenspace soils. However, the context of the soil was important, and in soils with no additions, C stocks were lower and nutrient dynamics did appear to be altered. Hypothesis ii was accepted as all sealed soils exhibited lower nutrient (nitrogen and phosphorus) stocks compared to greenspace soils.

Chapter 4 addressed **objective 3** by investigating the effect of sealing and anthropogenic additions on soil C persistence using a study of functional pools of soil organic matter. It tested the hypotheses that, (i) sealed soils will have less mineral-associated OC and therefore less persistent OC; and (ii) soils with anthropogenic additions will have more mineral-associated OC and therefore more persistent OC in both sealed and greenspace soils. Hypothesis i was accepted, as sealed soils had less MOAM OC and therefore less persistent microbial-derived OC. Hypothesis ii, however, was rejected in sealed and greenspace soils. In sealed soils, high OC due to the addition of black C did not contribute to greater OC in the MOAM pool, despite greater OC in the POM pool. This black C would add a stable form of OC to the soil though this would not be an ecologically-derived persistent form of OC. In greenspace soils, additions did not contribute to greater OC in the MAOM pool and conversely, may have had a detrimental effect as there was less N in the MAOM pool and less dissolved OC. To our knowledge, this study is the first to consider the persistence of soil C in sealed soils using functional SOM pools to understand C stability.

Chapter 5 addressed **objective 4** by investigating the effects of depth and soil history on soil C stocks. It tested the hypothesis that soil C and N stocks will decrease with soil depth. It also explored the influence of soil history and development on soil C and N storage. The hypothesis was rejected as the typical pattern of a decrease over depth was not seen in sealed soils, while soil history was found to have a greater influence on soil C and N than depth. Soil history was found to play an important role in sealed soils as a result of former land uses and soil treatment over time, and resulting cultural layers and losses of soil through development.

This thesis has addressed various knowledge gaps at different scales by meeting these objectives. The key findings from these chapters and consideration of the hypotheses set out above are summarised in Figure 6.1, and the findings from across the whole thesis are set out in section 6.2.



1. Sealed soils are not always depleted of carbon. For example, sealed anthropogenic soils had additions of black carbon (char) which led to larger carbon stores than in greenspace soils.
2. Sealed soils had lower nutrient (nitrogen and phosphorus) stocks than greenspace soils.
3. Anthropogenic additions in sealed soils may enable more nutrient cycling than in sealed soils without additions, likely due to improved conditions such as more oxygen or water availability.
4. Sealed soils had less persistent mineral-associated (and microbially-derived) organic carbon than greenspace soils.
5. Anthropogenic additions in sealed soils did not lead to more mineral-associated organic carbon despite the presence of black carbon.
6. Anthropogenic additions in greenspace soils did not lead to more mineral-associated organic carbon, and may have been detrimental to nitrogen cycling.
7. The typical decrease usually seen in carbon and nitrogen over depth was not seen in deeper sealed soils due to cultural layers and the influence of soil history.

Figure 6.1: Summary of key findings on the effects of soil sealing on carbon and nutrients.

6.2 Summary of findings across the thesis

Several key findings have emerged across this thesis which contribute to our understanding of carbon storage in urban soils.

6.2.1 Sealed soils are not always depleted of C

Across this thesis, all soil survey data has illustrated that sealed soils were not depleted of C and, in some cases, provided high soil C stocks that were comparable to, or greater than, C stocks in greenspace soils. This was seen in chapters 3 and 4 where sealed soils had high C stores as a result of legacy black C additions to the soil and exhibited C stocks of 8.06 ± 4.65 kg C m^{-2} and 3.30 ± 2.15 kg OC m^{-2}, while greenspace soils exhibited stocks of 4.92 ± 1.11 kg C m^{-2} and 3.25 ± 0.80 kg C m^{-2}. High soil C storage was also seen in chapter 5 where cultural layers built up over hundreds of years of urbanisation contributed to C-rich soils, with C stocks ranging between 24-28 kg C m^{-2} over 1 m depth profiles. This contrasts with the majority of research into soil sealing, and while there are few studies, most indicate that sealing leads to a depletion of soil C (Raciti et al., 2012; Wei et al., 2014a; Wei et al., 2014b; Majidzadeh et al., 2017; Majidzadeh et al., 2018; Pereira et al., 2021). This is largely attributed to the loss of topsoil during construction which contains large amounts of OC, and the lack of plant roots, root exudates or leaf litter returned to the soil. However, a small number of recent studies have found surprisingly large soil C stores under sealed surfaces and have challenged the assumption that sealed soil cannot provide the function of C storage. In Leicester, New York and Seoul, sealed soils had no less C stock than greenspace soils when comparing deeper soils (Edmondson et al., 2012; Cambou et al., 2018; Bae and Ryu, 2020). This thesis supports these alternative studies and has shown that sealed soils are not depleted in C but can, in fact, have high soil C stores. It has also been suggested that sealed soils could be overlooked as hotspots of soil C in urban environments (Bae and Ryu, 2020) and that sealing may provide stores of soil C that are protected from decomposition (Vasenev and Kuzyakov, 2018).

The context and age of the urban area is important when inferring findings in urban soil studies. It is worth noting that some previous studies that found sealed soils to be depleted of C were undertaken in cities younger than Manchester, or in areas that were formerly greenspaces and were sealed primarily for the purpose of the study. Therefore, these scenarios are likely to lead to notably different outcomes than those studied here. In this thesis, soils under sealed surfaces are likely to have had longer and more complex development histories, with longer to accumulate cultural layers and artefacts such as black C.

6.2.2 *High sealed soil C stores may be vulnerable to losses*

While this thesis found high C stores in sealed soils, there are various pathways that make them vulnerable to losses.

6.2.2.1 *Loss of legacy black C and lack of OC replenishment*

Legacy OC stores in sealed soils could be easily lost by removal during construction and would likely not be replenished by ecologically-derived OC. Chapter 3 observed a legacy C store due to the use of coal char, or black C, added into sealed soils during road building throughout the 19th and 20th centuries. However, chapter 4 found that despite high OC in the particulate organic matter (POM) due to the black C, OC was depleted in the mineral-associated organic matter (MAOM), the functional pool that is largely ecologically-derived and provides the most persistent C store (Lavallee et al., 2020).

Black C, in the form of coal char, was a common waste material from industry and power generation during the 19th and 20th centuries and was used extensively in road foundations in the UK and USA (MacBride, 2013). Coal tar was also used in road construction in the UK from the mid-1800s until the late 1980s when roads became primarily bitumen. Due to the carcinogenic properties of coal tar and the ability of tar products to migrate through the layers of road foundations, the Environment Agencies of the UK consider all road planings and waste arisings to be hazardous and require their safe disposal (Transport Scotland, 2018). This limits the re-use of black C enriched soils under sealed surfaces and results in their loss during roadworks or redevelopment. Contemporary road building uses recycled stone and rubble materials as fill for foundations, and while this might contribute some anthropogenic and inorganic C to the soil, the amount of black C is likely to be much lower as it is now a less common waste material.

Microbial processes are severely limited in sealed soils (Zhao et al., 2012; Wei et al., 2013; Wei et al., 2014b; Piotrowska-Długosz and Charzyński, 2015; Pereira et al., 2021) and as a result it is likely that the processes leading to long-term soil OC storage are also limited. The MAOM pool is mostly comprised of microbially-derived products and, as such, it is strongly linked to microbially activity (Kleber et al., 2015; Cotrufo et al., 2019). Sealing depleted OC in the MAOM pool whether there were additions of black C or not, indicating that black C did not contribute to microbial activity or microbial products in sealed soils. It can therefore be determined that the lack of microbial activity in the sealed soils will result in a lack of microbial OC contribution to the MAOM pool. The burial of black C in sealed soils may serve as a mechanism for its persistence (Abney and Berhe, 2018). However, any disruption or removal of soil containing black C during development would cause the loss of the C store, with little or no replenishment from microbially-derived OC. In addition, it could also allow the black C to be vulnerable to priming, as recent studies have suggested that black C can be decomposed faster than previously thought (Hilscher et al., 2009; Nguyen et al., 2010; Zimmerman, 2010; Bird et al., 2015).

6.2.2.2 *Loss of cultural layers due to urban development*

Urban development may pose a threat to C-rich cultural layers in urban soils. Chapter 5 illustrated deep urban soil profiles with high soil C stores and a long history of urban development. The accumulation of materials added to the soil over years of human activities led to the creation of cultural layers, features that are known to be enriched in C (Alexandrovskaya and Alexandrovskiy, 2000; Vasenev et al., 2013; Vasenev and

Kuzyakov, 2018; Bae and Ryu, 2020). Chapter 5 also illustrated how disturbance and redevelopment can lead to losses of these cultural layers and the C store they provide. Their removal may lead to replacement with lower C content fill materials, while the C-rich soils may be disposed of in landfill. This problem is exacerbated in contemporary construction, and the loss of intermediate horizons from urban soils has been observed on a large scale across numerous cities in the USA (Herrmann et al., 2018). In the UK, a large volume of uncontaminated soil from construction sites is disposed of every year, with 29.5 million tonnes sent to landfill in 2018 (Defra, 2021). As a comparison, the amount of soil lost to erosion in England and Wales is approximately 2.9 million tonnes per year (Graves et al., 2015). This could represent a wider problem that is under-researched and rarely considered in practice. Current construction practice does not take account of the C losses that arise due to construction and redevelopment, and this may be leading to a loss of valuable C-rich soils from cities.

6.2.3 Soil history drives heterogeneity and determines soil ecosystem services

This thesis showed consistently that urban, and particularly sealed, soils are highly heterogeneous. Soil history appeared to drive this heterogeneity and played a key role in determining the soil properties and soil functions. This thesis illustrates that we cannot expect urban soils with varied soil histories to all provide the same ecosystem services and functions.

Soil history determines the anthropogenic materials, artefacts and cultural layers found within urban soils. This thesis found that the effects of artefacts varied across sealed and greenspace soils, and the outcome of soil histories and artefacts was not always consistent. Chapters 3 and 4 showed that artefacts in sealed soils notably altered C and N stocks, soil OC persistence and mineral N dynamics. In greenspaces, artefacts led to lower N stock, lower N in the mineral-associated OM, and lower dissolved OC, likely a result of the additions limiting decomposition and potentially posing a threat to ecological processes and long-term OC storage in greenspaces. Chapter 5 illustrated that cultural layers and artefacts in sealed soils led to high C values and variable distribution over depth of C and N stocks, extractable OC and inorganic C, and pH. Thus, the history and heterogeneity of urban soils needs to be considered to understand urban soil C storage.

It is only recently that the importance of considering the legacy effects of soil history has been emphasised (Ziter and Turner, 2018; Delbecque et al., 2022); likely a result of urban soil research being a relatively young field. The heterogeneity of urban soil has long been discussed by soil scientists (Craul, 1985; Bullock and Gregory, 1991; Lehmann and Stahr, 2007; Pouyat et al., 2007; Raciti et al., 2011; Vasenev et al., 2013; Pouyat et al., 2020; Cambou et al., 2021), however, soil history is not often emphasised as the driving factor behind this soil heterogeneity, and many studies instead focus on current land use, vegetation cover, soil management or urbanisation gradients as key controlling factors (Martinová et al., 2016; Weissert et al., 2016; Tresch et al., 2019a; Canedoli et al., 2020). This has implications for our understanding of urban soil ecosystem services as it will not be possible to fully understand and predict their provision if we do not consider the soil history and context. This also reflects a key finding from chapter 2, that cultural ecosystem services are rarely considered in urban soils. Cultural layers accumulate over time in urban soils, and not only do they provide a large C store, they also provide a record of former human settlement and economic development of a city (Alexandrovskaya and Alexandrovskiy, 2000; Mazurek et al., 2016). The lack of consideration of soil history limits our understanding of urban soil, its heterogeneity, and its ability to provide ecosystem services. This lack of understanding contributes to the poor protections currently in place for urban soil in planning policy, and highlights the need for greater understanding of urban soils, better urban soil mapping, and more detailed urban soil surveys in development projects.

6.2.3.1 *Heterogeneity or homogeneity due to urbanisation*

In contrast to the acknowledged heterogeneity of urban soils, there is a small body of literature supporting an 'urban ecosystem convergence hypothesis', such that ecosystem properties in urban areas converge to similar levels within and among cities (Groffman et al., 2014; Pouyat et al., 2015; Schmidt et al., 2017; Herrmann et al., 2020). It is suggested that this homogenisation may have effects on soil C sequestration, fungal diversity and microclimate and may be driven by human actions at the household scale (Groffman et al., 2014). The theory has been tested on urban soil properties in only three studies, one of which showed urbanisation led to convergence of soil C and soil texture away from more heterogeneous approximated pre-urban conditions (Herrmann et al., 2020); and one that found soil C and N were less variable in residential yards compared to nearby natural reference sites (Trammell et al., 2020). The third study showed partial support for the hypothesis, with properties that are affected by anthropogenic and biogenic processes converging, such as soil OC and N (Pouyat et al., 2015). These studies illustrated convergence effects at the continental scale in cities across different climatic regions of the USA. While this thesis did not compare urban to natural or pre-urban soils, it has found that heterogeneity within the city studied, Manchester (UK), was a major controlling factor in urban soil properties. A possible reason for this difference is the age of Manchester and the long development history of the area in comparison to many US cities; while another likely reason may be that this pattern of convergence is not seen in sealed soils. These studies only considered greenspace soils, and therefore, it would be beneficial for future research to investigate whether this convergence may be occurring in UK sealed soils by making a comparison against non-urban or pre-urban approximations.

6.2.3.2 *Soil history and Technosols*

Soil heterogeneity in historical Technosols may be overlooked in current Technosol research and needs to be considered to understand urban soil functioning. The soils studied in chapters 4, 5 and 6 provided examples of historical Technosols as a result of being sealed and containing artefacts. These chapters highlighted the heterogeneity of anthropogenic additions to the soil and the effects that they had on soil C and N stores. Chapter 2 highlighted the use of Technosols as an important driver of future change for cities, and recommended further research into their study in relation to soil functions.

Technosols are defined in the World Reference Base as containing at least 20 % artefacts in the top 100 cm, or being sealed with a hard impermeable material or geomembrane at the soil surface (FAO, 2015). They are being increasingly studied, in particular, 'constructed Technosols', which are soils that are made to mimic natural soils while providing a use for waste materials (Fabbri et al., 2021). Studies often focus on measuring soil properties in constructed Technosols rather than in historical or in-situ Technosols that have formed unintentionally or over long periods of time (Scalenghe and Ferraris, 2009; Rees et al., 2019; Deeb et al., 2020; Ivashchenko et al., 2021). The Technosol literature is growing rapidly and provides vital information on this relatively young and important topic. Therefore, the consideration of historical Technosols needs to be joined up with research on contemporary and constructed Technosols, particularly in light of research into the circular economy and reuse of materials and resources including soils and development (Breure et al., 2018; Fabbri et al., 2021). While it will make a valuable contribution to urban soil research, it is important to note that findings on constructed Technosols will likely not be representative of historical Technosols or cultural layers, and this needs to be acknowledged when considering soil functions in cities with a long settlement history.

6.3 Research output and impact

At the time of submitting this thesis, two chapters had been published in peer-reviewed journals. Chapter 2 was published in *Geoderma* and was highlighted as Editor's Choice for March 2021. Following this, the findings of this chapter were communicated in an article for *The Conversation* website, with the aim of promoting urban soils and the ESs they provide. Chapter 3 was published in the journal *SOIL* in 2021.

The work in this thesis facilitated inputs to an Impact Acceleration Account grant at Lancaster University. This work will lead to a guidance document to manage soils more sustainably on construction sites, and is engaging professionals across the planning, design and construction industries. Therefore, findings from this thesis will lead to more informed guidance for maintaining soil functions during construction.

6.4 Conclusions

This thesis has contributed new insights into our understanding of urban soil ESs, the effects of sealing on urban soil C and nutrients and the influence of anthropogenic additions on soil functions. It identified urban soil multifunctionality as a gap in the research literature and highlighted this as a key mechanism to enable soil functions to be recognised in urban planning and management. Legacy soil C stores were identified in sealed soils as a result of historical black C additions, and in some cases, provided C stocks that were comparable to, or greater than, C stocks in greenspace soils. This provided a valuable addition to the small body of work on sealed soil OC. Analysis of functional SOM pools revealed that the legacy C store contributed only to the particulate pool but did not contribute to the microbially-derived mineral-associated pool; therefore this legacy C is vulnerable to loss during construction with no replenishment from ecological processes. This was the first time this fractionation method had been used on sealed soils and it provides valuable insights into the depletion of long-term persistent mineral-bound OC in sealed soils. In greenspace soils, it illustrated that artefacts added to the soil could potentially limit ecological processes and negatively impact long-term soil OC storage. A study of depth profiles under sealed surfaces revealed that soil history was a major control on soil OC and N rather than depth. This highlighted the importance of soil history and reflected findings from across this thesis that soil history is a driver of urban soil heterogeneity and determines soil functions and ecosystem services. In light of this, soil history and its heterogeneity needs to be considered in urban soil mapping, urban planning and construction.

6.5 Future research

This thesis advances our knowledge of urban soil C storage across sealed and greenspace soils. However, it has also highlighted several areas for future interesting research:

1. **Urban soil multifunctionality** – Chapter 2 found that urban soil research focuses on supporting processes and functions and a few selected regulation ESs. However, there is a need for research that addresses a wider range of ESs together to illustrate the value of urban soil and shed light on the trade-offs that need to be considered for urban soil management. In particular, further studies of water dynamics, urban food growing and cultural services would benefit from linking up to the wider ESs framework and research literature.
2. **Ecological processes in sealed soils** – In chapter 4, it is likely that black C did not contribute to microbial processes and the long-term persistent store of OC in sealed soil. Biochar is increasingly being used in urban soils, and therefore, future studies into its use in sealed soils would be beneficial. Future studies into the microbial processes occurring in sealed soils would help to build greater understanding of OC persistence, potential interactions with biochar and outcomes for OC storage. This would enable better long-term management of soil OC during construction and development projects.
3. **The effect of anthropogenic additions in greenspace soils** – Chapter 4 illustrated that greenspace soils with additions had lower N stocks, lower N in the mineral-associated OM pool and lower dissolved OC. This was likely due to a reduction in decomposition in these soils and indicates that anthropogenic additions to greenspace soils may pose a threat to important soil functions. Future studies should investigate this further by considering soil functions and the effect of anthropogenic additions on primary productivity in urban greenspaces.
4. **Enhanced weathering in urban soils** – Previous work has found that urban soils can be a site of enhanced weathering which leads to atmospheric CO_2

absorption and storage as carbonate. Chapters 3, 4 and 5 found high inorganic C in both sealed and greenspace soils, a feature which is common is urban soils. This may be a result of high Ca^+ ions in urban soils due to Ca bearing materials added to soils such as concrete and limestone. While a small body of work considers this for urban soils on brownfield sites, future work should also extend this to both greenspace soils and sealed soils where the process is also likely occurring and may contribute to total C stores.

5. **Losses of urban soil C** – This thesis has illustrated across chapters 3, 4 and 5 that sealed soils are not depleted of soil C, and in some cases provide high stores of legacy C and historical cultural layers rich in C. Current development practices do not value soil as a resource and much urban soil on construction sites is lost to landfill or severely damaged. To address this, it would be beneficial to calculate the C cost of construction projects and illustrate the losses of this vital resource, a quantification that is currently not considered in planning or C assessments for new developments.

7. References

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8. Appendix 1: Chapter 2 – Supplementary material

Table 8.1: The list of supporting processes and ecosystem services as set out in the framework by Dominati et al. (2010), and an explanation of how papers were categorised in the manuscript. Definitions of supporting processes and ecosystem services given by Dominati et al. (2010) have been abbreviated.

Dominati et al. (2010) soil ES framework			**Categorisation used in this review**
Category	**Process or ES**	**Abbreviated Definition**	**Papers that measure…**
Supporting processes	Nutrient cycling	The processes by which a chemical element moves through both the biotic and abiotic compartments of soils. Nutrient cycles are a way to conceptualise the transformations of elements in a soil.	Concentrations, transformations and availability of soil nutrients, such as carbon, nitrogen and phosphorus.
	Water cycling	The physical processes enabling water to enter soils, be stored and released. Soil moisture is the driver of many chemical and biological processes and is therefore essential in soil development and functioning.	Soil hydrological processes and quantities, such as soil moisture, infiltration and water movement through soil.
	Soil biological activity	Soils provide habitat to a great diversity of species, enabling them to function and develop. In return, the activity and diversity of soil biota are essential to soil structure, nutrient cycling, and detoxification.	Soil biological properties, such as type and quantity of soil fauna; and soil biological activities, such as respiration or decomposition.

Provisioning services	Food, wood and fibre	Humans use a great variety of plants for a diversity of purposes (food, building, energy, fibre, medicines). By enabling plants to grow, soils provide a service to humans.	The provision of plants for food, wood or fibre.
	Physical support	Soils form the surface of the earth and represent the physical base on which animals, humans and infrastructures stand. Even an otherwise unproductive soil may provide physical support to human infrastructures.	The physical support provided by soil to urban infrastructure.
	Raw materials	Soils can be source of raw materials like, for example, peat for fuel and clay for potting. These materials stocks are the source of the service.	The provision of raw materials.
Regulating services	Flood mitigation	Soils have the capacity to store and retain quantities of water and therefore can mitigate and lessen the impacts of extreme climatic events and limit flooding. Soil structure and more precisely macroporosity, as well as processes like infiltration and drainage will impact on this service.	The capacity of soil to alleviate flooding, such as storage and retention of water, run-off volume, structure and macroporosity, impermeable surfaces and SUDs.
	Filtering of nutrients	If the solutes present in soil (e.g. nitrates, phosphates) are leached, they can become a contaminant in aquatic ecosystems (e.g. eutrophication) and a threat to human health (e.g. nitrate in	The ability of soil to filter nutrients, such as filtration and retention capacity, leaching and water quality.

		drinking water). Soils have the ability to absorb and retain solutes, therefore avoiding their release into water.	
	Biological control of pests and diseases	By providing habitat to beneficial species, soils can support plant growth and control the proliferation of pests (crops, animals or humans' pests) and harmful disease vectors (e.g. viruses, bacteria). Soil conditions determine the quality of the soil habitat and thereby select the type of organisms present.	The ability of soil to provide a habitat for beneficial species that prevent pests and disease.
	Recycling of wastes and detoxification	Soils can self-detoxify and recycle wastes. Soil biota degrades and decomposes dead organic matter into more simple forms that organisms can reuse. Soils can also absorb (physically) or destroy chemical compounds that can be harmful to humans, or organisms useful to humans.	The recycling and decomposition of wastes and contaminants, such as remediation, detoxification, degradation and content of contaminants.
	Carbon storage and regulation of greenhouse gas emissions	Soils play an important role in regulating many atmospheric constituents, therefore impacting on air quality. Perhaps most important is the ability of soils to store carbon as stable organic matter which is a non-negligible benefit when talking about off-setting greenhouse gases emissions.	The quantity of carbon stored in soil or emission of greenhouses gases, such as soil carbon content or stocks, carbon sequestration or greenhouse gas regulation.

Cultural services	Culture	Spirituality, knowledge, sense of place, aesthetics… The point here is not to detail all the cultural services provided by soils but to acknowledge that these services, even if almost always forgotten, are of tremendous consequence.	Cultural services provided by urban soils.

9. Appendix 2: Chapter 3 – Table of Results

Table 9.1: Urban soil measurements showing the mean (±SD). Different letters indicate a statistically significant difference between soil categories at the 0.05 level.

Measurement	**Sealed undisturbed (n=22)**		**Sealed anthropogenic (n=14)**		**Greenspace soil (n=32)**		***P* value**
Soil Properties							
pH	8.03a	(±0.63)	8.44a	(±0.62)	6.82b	(±0.98)	<0.001
Bulk density (g cm^3)	1.50a	(±0.13)	1.32b	(±0.18)	0.86c	(±0.14)	<0.001
Soil moisture content (%)	21.38ab	(±4.21)	18.63a	(±5.84)	27.17b	(±11.63)	0.006
Soil Carbon							
Total carbon stock at 0-10 cm (kg m^{-2})	3.10a	(±1.45)	8.06b	(±4.65)	4.92c	(±1.11)	<0.001
Soil organic matter (%)	5.00a	(±1.29)	7.06a	(±3.20)	11.12b	(±2.76)	<0.001
Extractable Organic Carbon (mg kg^{-1})	132.04a	(±79.88)	107.92a	(±41.11)	602.84b	(±204.02)	<0.001
Extractable Inorganic Carbon (mg kg^{-1})	34.90a	(±24.35)	34.31a	(±32.20)	9.79b	(±10.25)	<0.001
Nutrients							
Total N stock at 10 cm (g m^{-2})	81.20a	(±22.47)	90.11a	(±28.38)	115.44b	(±26.49)	<0.001
Ammonium (mg kg^{-1})	11.06a	(±15.52)	2.53a	(±4.88)	6.10a	(±11.74)	0.100
Nitrate (mg kg^{-1})	0.70a	(±0.65)	2.95ab	(±4.11)	7.82b	(±12.30)	<0.001
Total P stock at 10 cm (g m^{-2})	39.62a	(±20.91)	39.42a	(±23.68)	62.62b	(±36.67)	0.003
Stoichiometry							
C:N ratio	37.29a	(±12.44)	86.84b	(±40.45)	43.10a	(±6.99)	<0.001
C:P ratio	83.81a	(±31.35)	248.69b	(±208.24)	94.19a	(±37.23)	<0.001
N:P ratio	2.42a	(±1.04)	2.81a	(±1.27)	2.25a	(±1.06)	0.300

10. Appendix 3: Chapter 3 – Supplementary material

Sealed Undisturbed soils

Sealed Anthropogenic soils

a) Tarmac surface, under which older road cobbles are visible. No significant road foundations visible under cobbles. Clay rich soil visible through the profile.

b) Paving surface, under which there is a layer of sharp sand and some aggregate paving foundation material. Soil underneath this material is at 60 cm depth.

c) Tarmac road and pavement surface. Foundation materials are visible to a depth of 10 cm, followed by darker materials, possibly ash or charcoal. This is followed by a clay layer mixed with aggregate materials, followed by the clay rich soil layer below at 70 cm depth.

d) Cement surface layer underlain by a thin layer of aggregate then a thicker layer of limestone gravel. Under this, finer aggregate is mixed in with soil to approximately 75 cm depth.

Figure 10.1: Examples of sealed soil profiles with descriptions, showing (a-b) sealed undisturbed (SU) soils; and (c-d) sealed anthropogenic (SA) soils.

11. Appendix 4: Urban soil microbial community and microbial-related carbon storage are severely limited by sealing

In addition to the chapters presented in this thesis, I also contributed to the following paper which was published in the *Journal of Soils and Sediments* in 2021. With permission from co-authors, I include it here.

I hereby agree with the above statement:

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SOILS, SEC 5 • SOIL AND LANDSCAPE ECOLOGY • RESEARCH ARTICLE

Urban soil microbial community and microbial-related carbon storage are severely limited by sealing

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Abstract

Purpose Urbanisation causes changes in land use, from natural or rural to urban, leading to the sealing of soil and the replacement of vegetation by buildings, roads and pavements. The sealing process impacts soil properties and services and can lead to negative consequences for microbial attributes and processes in soil. At present, information about the microbial community following soil sealing is limited. As such, we investigated how changes in soil physical and chemical properties caused by sealing affect the soil microbial community and soil ecosystem services.

Material and methods Soils were sampled beneath impervious pavements (sealed) and from adjacent pervious greenspace areas (unsealed). Soil properties (total C, total N, C:N ratio and water content) and microbial attributes (microbial biomass C, N-mineralisation and phospholipid fatty acids—PLFA) were measured and correlated.

Results and discussion A reduction of total C, total N, and water content were observed in sealed soil, whilst the C:N ratio increased. Sealed soil also presented a reduction in microbial attributes, with low N-mineralisation revealing suppressed microbial activity. PLFA data presented positive correlations with total C, total N and water content, suggesting that the microbial community may be reduced in sealed soil as a response to soil properties. Furthermore, fungal:bacterial and gram-positive:gram-negative bacterial ratios were lower in sealed soil indicating degradation in C sequestration and a consequential effect on C storage.

Conclusions Sealing causes notable changes in soil properties leading to subsequent impacts upon the microbial community and the reduction of microbial activity and soil C storage potential.

Keywords Urban soil · Soil sealing · Impervious surfaces · Microbial biomass · N-mineralisation · PLFA · Soil carbon · Carbon storage

1 Introduction

Urbanisation causes considerable impacts on soil properties and services (Yan et al. 2015, 2016). Changes in land use from natural and rural to urban are associated with the replacement of vegetation by buildings, roads and pavements (Edmondson et al. 2012; Yan et al. 2016). The high degree of impermeable surfaces in cities has many negative consequences for the environment and the services it provides, particularly those provided by soil (Morgenroth et al. 2013; Wei et al. 2013; Piotrowska-Długosz and Charzyński 2015; Ziter and Turner 2018; Kelleher et al. 2020).

Carbon (C) storage is an important ecosystem service provide by soil in urban areas, with vegetation biomass inputs and soil organic carbon (SOC) being key components of overall C storage (Edmondson et al. 2012; Ziter and Turner 2018). Soil sealing due to urbanisation leads to the removal of plants and topsoil during the paving and construction process. This results not only in large losses of C stocks from urban soil (Wei et al. 2014) but also alters soil C dynamics, typically leading to a loss of SOC (Majidzadeh et al. 2018). Previous soil C inventories suggested that urban soil provides very little or no soil C storage (Bradley et al. 2005). However, more recently, significant amounts of soil C have been reported in urban areas, in soils of greenspaces and beneath sealed surfaces of pavements and

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houses (Edmondson et al. 2012; Wei et al. 2014; Majidzadeh et al. 2017; Yan et al. 2016; Hu et al. 2018; Vasenev et al. 2018). As such, urban soil C and the dynamics of C storage are receiving increasing attention in research literature.

Many other key ecosystem services and soil properties are affected by soil sealing. Water infiltration is prevented or reduced, changing surface runoff patterns and seasonal dynamics of soil water content (Majidzadeh et al. 2018; Hu et al. 2020; Kelleher et al. 2020). Paving materials can act as a reservoir for contaminants such as heavy metals (Hu et al. 2018) and polycyclic aromatic hydrocarbons (Li et al. 2020), and soil temperatures can be increased (Chen et al. 2016, 2017). Gas exchange between the soil and atmosphere is reduced which can lead to higher CO_2 concentrations in sealed soil and increased CO_2 flux rate near pavement edges (Wu et al. 2016; Fini et al. 2017). Additionally, soil nutrient content can be altered, with sealed soils exhibiting increased calcium, potassium, sodium and phosphorous and decreased aluminium, iron, magnesium and nitrogen (Zhao et al. 2012; Morgenroth et al. 2013; Hu et al. 2018; Majidzadeh et al. 2018). The severe decrease in nitrogen (N) can lead to very high CN ratios in sealed soils, despite the concurrent loss of soil C (Zhao et al. 2012; Hu et al. 2018).

These changes to the soil environment also affect soil microbes, which may impact the microbial processes and activities that underpin many important soil services (Zhao et al. 2012). Whilst sealed soils remain largely understudied, a small number of studies have observed that sealing can lead to a decrease in microbial biomass C, microbial biomass N, enzyme activities and respiration potential (Zhao et al. 2012; Wei et al. 2013; Piotrowska and Charzynski 2015), as well as a decrease in N-mineralisation potential (Zhao et al. 2012; Majidzadeh et al. 2018). Similarly, sealing has led to changes in bacterial communities, with a reduction in alpha diversity and a distinct community found in sealed soil when compared with unsealed soil (Hu et al. 2018; Yu et al. 2019). Research has shown that sealing has a negative effect on urban soil microbial attributes and bacterial communities, although little is known about the dynamics of both bacterial and fungal communities and their contribution to the soil microbial community in sealed soils. Furthermore, there is a gap in knowledge into what these altered bacterial and fungal dynamics mean for important soil ecosystem services such as nutrient cycling and C storage within sealed soils. Fungal:bacterial dominance is considered an important factor in C sequestration (Strickland and Rousk 2010), and the ratio between gram-positive:gram-negative bacteria provides insight into the stability or recalcitrance of C in the soil (Fanin et al. 2019). At present, these dynamics have not been studied in sealed soil, and therefore, the implications for soil C storage across the urban landscape are currently unknown.

In this paper, we investigate how changes to soil physical and chemical properties caused by sealing affect the microbial community and microbial attributes. The city of Lancaster (UK) and surrounding urban areas were used as a study site. We measure the soil properties (total C, total N, C:N ratio and water content) and microbial attributes (microbial biomass C, phospholipid fatty acids and N-mineralisation) to make a comparison across sealed and unsealed soils. To our knowledge, we present the first investigation into bacterial and fungal dynamics in sealed soil using phospholipid fatty acid analysis and consider their contributions to the soil microbial community and consequences for important soil services. We hypothesise that (i) sealing leads to large changes in soil properties, and (ii) sealing leads to changes in microbial attributes, significantly altering community composition and reducing microbial activity. Measurements of soil total C, total N, C:N ratio and water content provided indicators of the impacts of sealing on soil properties (hypothesis 1). Microbial biomass C, phospholipid fatty acids and N-mineralisation were used as indicators of changes in microbial attributes, with biomass C and phospholipid fatty acids pointing to changes in community composition, and N-mineralisation to changes in microbial activity (hypothesis 2).

2 Materials and methods

2.1 Study area

The study area consisted of the medium-sized UK city of Lancaster and the surrounding urban areas (Fig. 1). The National Soil Map for England and Wales, accessed on the Soilscapes Viewer online (www.landis.org.uk), shows that, across much of Lancaster city, there are freely draining slightly acid loamy soils, whilst sampling sites in the surrounding areas tended to be on slowly permeable seasonally wet acid loamy and clayey soils.

2.2 Soil sampling

Sealed soils were collected from 25 roadwork sites where works had exposed the soil beneath pavements and roads. Sealing had occurred at different times in the past, and further research is still needed to determine if the time since sealing has an impact on the measured variables. Soil was collected from the top 10 cm of soil below the sealed surface and human-made layers. To allow a comparison between soils, an unsealed sample was collected from the nearest available greenspace after each sealed soil. Unsealed samples were collected from the top 10 cm of soil, primarily from grass-covered road verges, amenity greenspaces and residential gardens, with a distance ranging from 0.5 to 15 m of the respective sealed site. Approximately 500 g of both soils (50 samples) were collected with a trowel and were immediately returned to the lab for refrigeration prior to fresh soil tests.

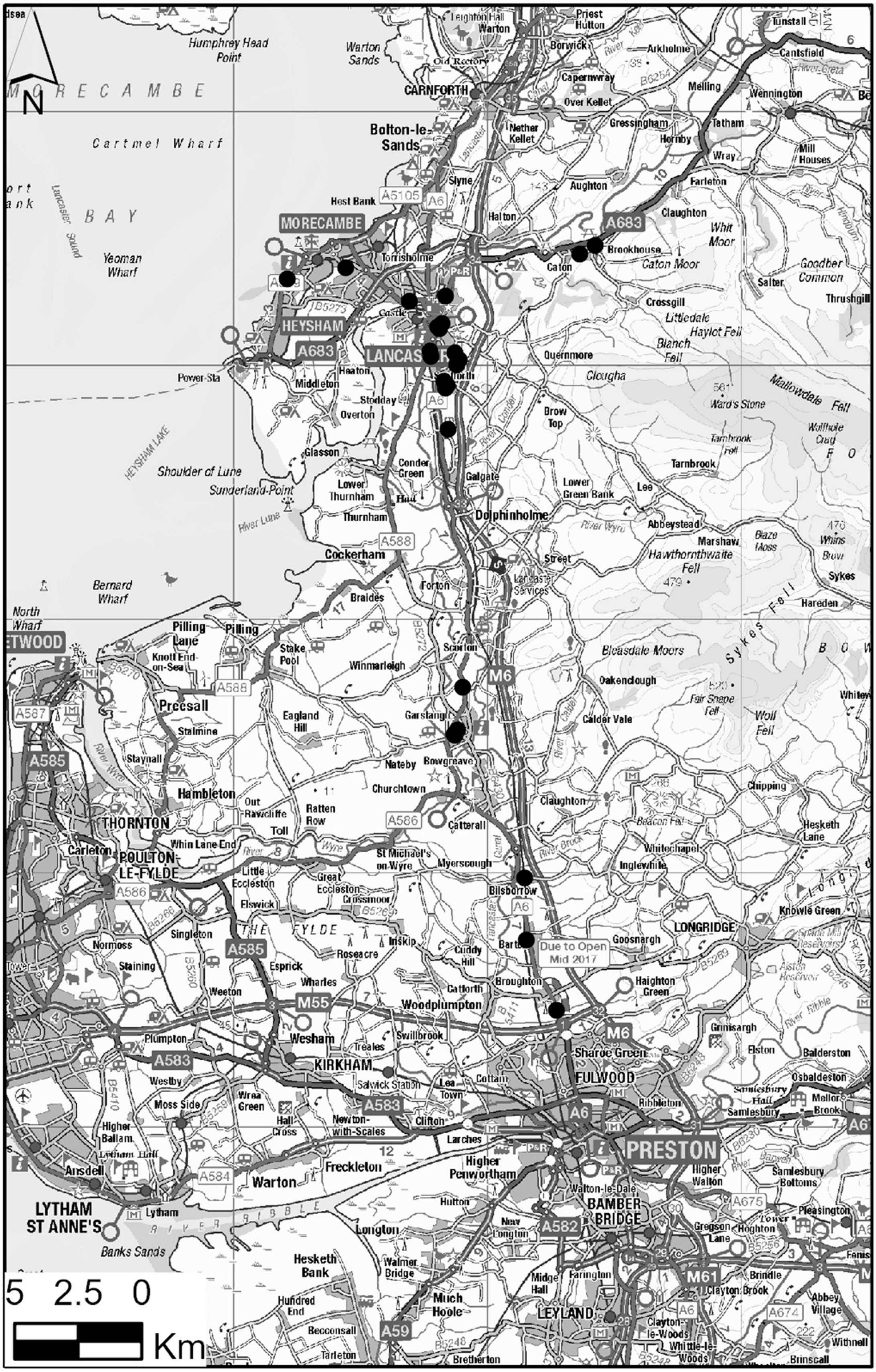

Fig. 1 Location of sampling sites, indicated on the map with black dots

2.3 Soil preparation and analysis

2.3.1 Soil properties and CN analysis

Soil water content was determined gravimetrically by drying the samples at 105 °C for 24 h. The dried sample was ball-milled to a powder and analysed for total C and total N using a dry combustion CN analyser (vario Max CN).

2.3.2 Microbial biomass C and N-mineralisation

Microbial biomass C (MBC) was determined using the chloroform fumigation-extraction method (Brookes et al. 1985; Vance et al. 1987). Two subsamples of 5 g of moisture adjusted soil were prepared for each sample, one fumigated with alcohol-free $CHCl_3$ for 24 h and one non-fumigated stored at 4 °C. After removal of the $CHCl_3$, both subsamples were extracted with 25 mL of K_2SO_4 (0.5 M) for 30 min. The filtrate was analysed for extracted C using a TOC analyser (Shimadzu TOC-L_{CPN} TN).

Soil potential N-mineralisation was measured before and after incubation. Subsamples were prepared for water saturation to determine moisture adjustments for each sample. The subsamples were placed in a funnel with Whatman no. 1 filter paper, wet with Milli-Q water and periodically rewet over a 2-h period. They were then covered with cling film and drained for 2 h, weighed and oven-dried at 105 °C for 24 h. They were reweighed, and moisture adjustments were calculated to 60% for each sample. For extractions, 5 g of moisture adjusted soil was put in an extraction bottle, covered with covered with polythene and incubated at 25 °C for 14 days. A second sample was extracted immediately. The incubated and non-incubated subsamples were extracted using KCl (1 M), and the filtrate was analysed for inorganic N using an autoanalyzer (Elementar Vario EL III). Potential N-mineralisation was calculated as the difference in inorganic N before and after incubation.

2.3.3 Phospholipid fatty acid analysis

Phospholipid fatty acid (PLFA) analysis was used to determine the overall microbial community composition and dominance. Soil subsamples were taken from soils previously stored at –80 °C and extracted for PLFA determination by gas chromatography (Vestle and White 1989; Willers et al. 2015). Microbial PLFA markers were identified and measured as per the method by Frostegård et al. (2011) to estimate the total and group-specific microbial marker biomass. The i15:0, a15:0, i16:0, a17:0 and i17:0 PLFA markers were used as gram-positive (GP) bacteria markers, and 16:1ω7, cy17:0, cis18:1ω7 and cy19:0 as gram-negative (GN) bacteria markers. Total bacteria were estimated from the sum of GP and GN bacteria, and 15:0 marker mass. Total fungi were measured using 18:1ω9 and 18:2ω6,9 as markers. The 16:1ω5 was used as a proxy measurement for arbuscular mycorrhizal (AM) fungi. Total PLFA expresses total microbial marker biomass and was estimated as the sum of total bacteria, total fungi, AM fungi and 16:0, 16:1ω7, br17:0, 17:1ω8, 17:0 7-methyl, 18:0, br18:0, 18:1ω5 and 19:1 markers. The fungal:bacterial and GP:GN ratios were calculated by dividing the respective biomarker masses.

2.4 Statistical analysis

Data were evaluated using R (version 4.0) on the software RStudio (version 1.1.463). Since only water content and total C in unsealed soil presented data with a normal distribution according to the Shapiro-Wilk test, the non-parametric Wilcoxon test was applied. Where microbial attributes presented values equal to zero, they were considered null values (Table 1), whilst some soil samples did not present detectable amounts of PLFA during gas chromatography and so were excluded from the analysis. Boxplots were constructed using the ggplot package, and statistical significance was presented to compare sealed and unsealed soils. The correlations between soil properties and microbial attributes were estimated using the Spearman's rank correlation (ggcorrplot package).

3 Results

Sealed soils exhibited consistently lower values than unsealed soils across all measured soil properties and microbial attributes, other than the C:N ratio (Table 1). Total C ($p = 0.0026$), total N ($p < 0.001$), and water content ($p < 0.001$) were all significantly lower in sealed soil than unsealed soil (Fig. 2a, b, and d), whilst the C:N ratio ($p = 0.023$) was higher in sealed soil (Fig. 2c). All microbial attributes exhibited significantly lower values in sealed soil than unsealed soil: MBC, N-mineralisation, total PLFA, total fungi, AM fungi, total bacteria, GP bacteria and GN bacteria presented $p < 0.001$; fungal:bacterial ratio presented $p = 0.019$; and GP:GN bacterial ratio presented $p = 0.0017$ (Figs. 3 and 4).

Significant correlations were observed between soil properties and microbial PLFA attributes; however, MBC and N-mineralisation potential showed no correlation with soil properties in this study (Table 2). In sealed soil, total bacteria had a strong and positive correlation with total N (rho = 0.63, p = 0.038) and water content (rho = 0.71, p = 0.015), GP bacteria a strong and positive correlation with total N (rho = 0.63, p = 0.038) and water content (rho = 0.71, p = 0.015), and GN bacteria a strong and positive correlation with total C (rho = 0.64, p = 0.032), total N (rho = 0.71, p = 0.015) and water content (rho = 0.79, p = 0.004). In unsealed soil, total PLFA, total fungi, total bacteria and GP bacteria presented moderate to strong positive correlations with total C (rho = 0.58, p =

Table 1 Descriptive statistics of soil properties and microbial attributes in sealed and unsealed soils

Variable groups	Variables	Pavement types	*n*	Null*	Min–max	Mean ± SE	CV* (%)
Soil properties	Total C g/Kg	Sealed	25	0	3.35–250.29	49.78 ± 10.67	107.12
		Unsealed	25	0	14.02–128.49	73.49 ± 5.33	36.27
	Total N g/Kg	Sealed	25	0	0.39–13.75	2.08 ± 0.587	141.03
		Unsealed	25	0	0.026–21.75	5.36 ± 0.79	73.24
	C:N ratio	Sealed	25	0	4.92–149.87	35.81 ± 7.13	99.57
		Unsealed	25	0	5.91–27.49	15.55 ± 1.11	35.62
	Water content g/g	Sealed	25	0	0.09–0.74	0.30 ± 0.03	54.09
		Unsealed	25	0	0.08–0.85	0.47 ± 0.03	32.83
Microbial attributes	MBC g/Kg	Sealed	25	7	0–47.85	6.11 ± 2.17	177.67
		Unsealed	25	0	1.99–58.59	19.79 ± 3.04	76.69
	Mineralisation g/Kg	Sealed	25	12	0–2.87	0.42 ± 0.15	178.14
		Unsealed	25	1	0–21.22	5.61 ± 1.03	92.12
	Total PLFA mg/Kg	Sealed	11	0	0.007–2.176	0.311 ± 0.198	211.30
		Unsealed	16	0	0.338–2.996	1.101 ± 0.164	59.61
	Fungi mg/Kg	Sealed	11	3	0–0.239	0.036 ± 0.021	197.03
		Unsealed	16	0	0.118–0.867	0.357 ± 0.050	56.33
	AM fungi mg/Kg	Sealed	11	8	0–0.019	0.003 ± 0.002	230.69
		Unsealed	16	0	0.008–0.146	0.062 ± 0.009	60.98
	Bacteria mg/Kg	Sealed	11	5	0–0.832	0.094 ± 0.075	263.19
		Unsealed	16	0	0.075–0.821	0.304 ± 0.045	58.83
	GP bacteria mg/Kg	Sealed	11	5	0–0.364	0.043 ± 0.033	249.92
		Unsealed	16	0	0.044–0.572	0.187 ± 0.032	68.37
	GN bacteria mg/Kg	Sealed	11	6	0–0.468	0.050 ± 0.042	277.50
		Unsealed	16	0	0.031–0.236	0.113 ± 0.013	45.08
	Fungal:bacterial ratio	Sealed	10	4	0–2.470	0.663 ± 0.284	135.20
		Unsealed	16	0	0.717–1.585	1.206 ± 0.062	20.57
	GP:GN bacterial ratio	Sealed	10	5	0–2.151	0.628 ± 0.237	119.30
		Unsealed	16	0	0.958–2.428	1.584 ± 0.104	26.16

n the number of values, *null* the number of null values, *min* the minimal value, *max* the maximal value, *SE* the standard error of the mean, *CV* the coefficient of variation

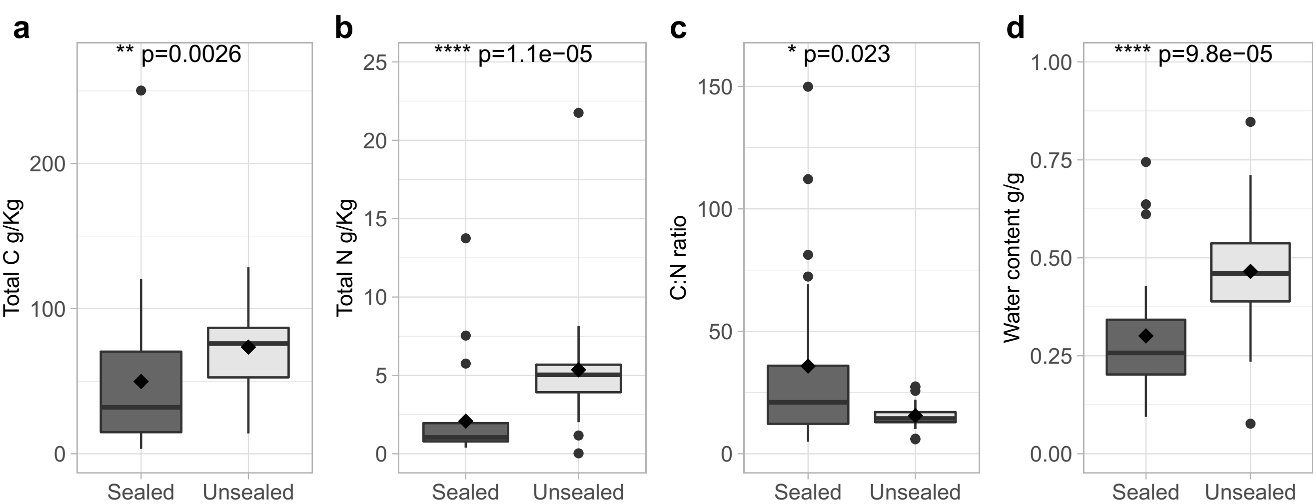

Fig. 2 Soil properties in sealed and unsealed soils. **a** Total C, **b** total N, **c** C:N ratio and **d** water content. A significant difference between sealed and unsealed soils was estimated by Wilcoxon test, with "****", "***", "**" and "*" indicating significance at $p < 0.0001$, $p < 0.001$, $p < 0.01$ and $p < 0.05$, respectively

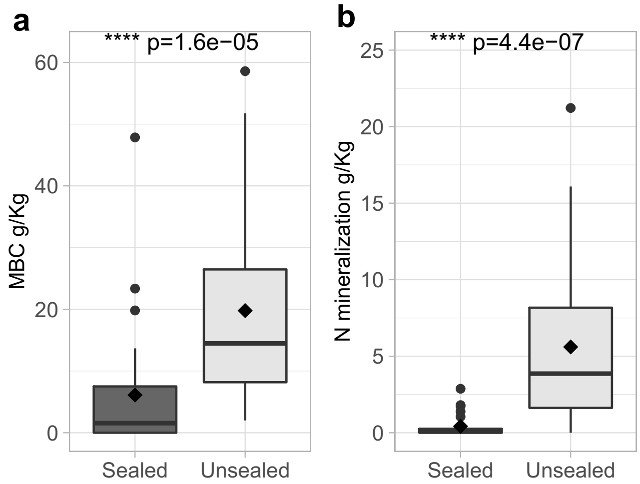

Fig. 3 Microbial biomass C (MBC) and N-mineralisation potential in sealed and unsealed soils. A significant difference between sealed and unsealed soils was estimated by Wilcoxon test, with "****" indicating significance at $p < 0.0001$

0.020; rho = 0.59, $p = 0.019$; rho = 0.56, $p = 0.025$ and rho = 0.52, $p = 0.042$, respectively); total N (rho = 0.62, $p = 0.012$; rho = 0.54, $p = 0.034$; rho = 0.68, $p = 0.005$; and rho = 0.69, $p = 0.004$, respectively); and water content (rho = 0.75, $p = 0.001$; rho = 0.75, $p = 0.001$; rho = 0.68, $p = 0.005$; and rho = 0.66, $p = 0.007$, respectively). GN bacteria had a strong positive correlation with total N (rho = 0.61, $p = 0.015$) and water content (rho = 0.65, $p = 0.008$), and the GP:GN bacterial ratio showed a moderate positive correlation with total C (rho = 0.52, $p = 0.040$).

4 Discussion

In contrasting soil samples from sealed and unsealed areas, we observed that sealing affects soil properties, reduces the microbial community and limits microbial processes, changes which may disrupt important soil ecosystem services. Soil

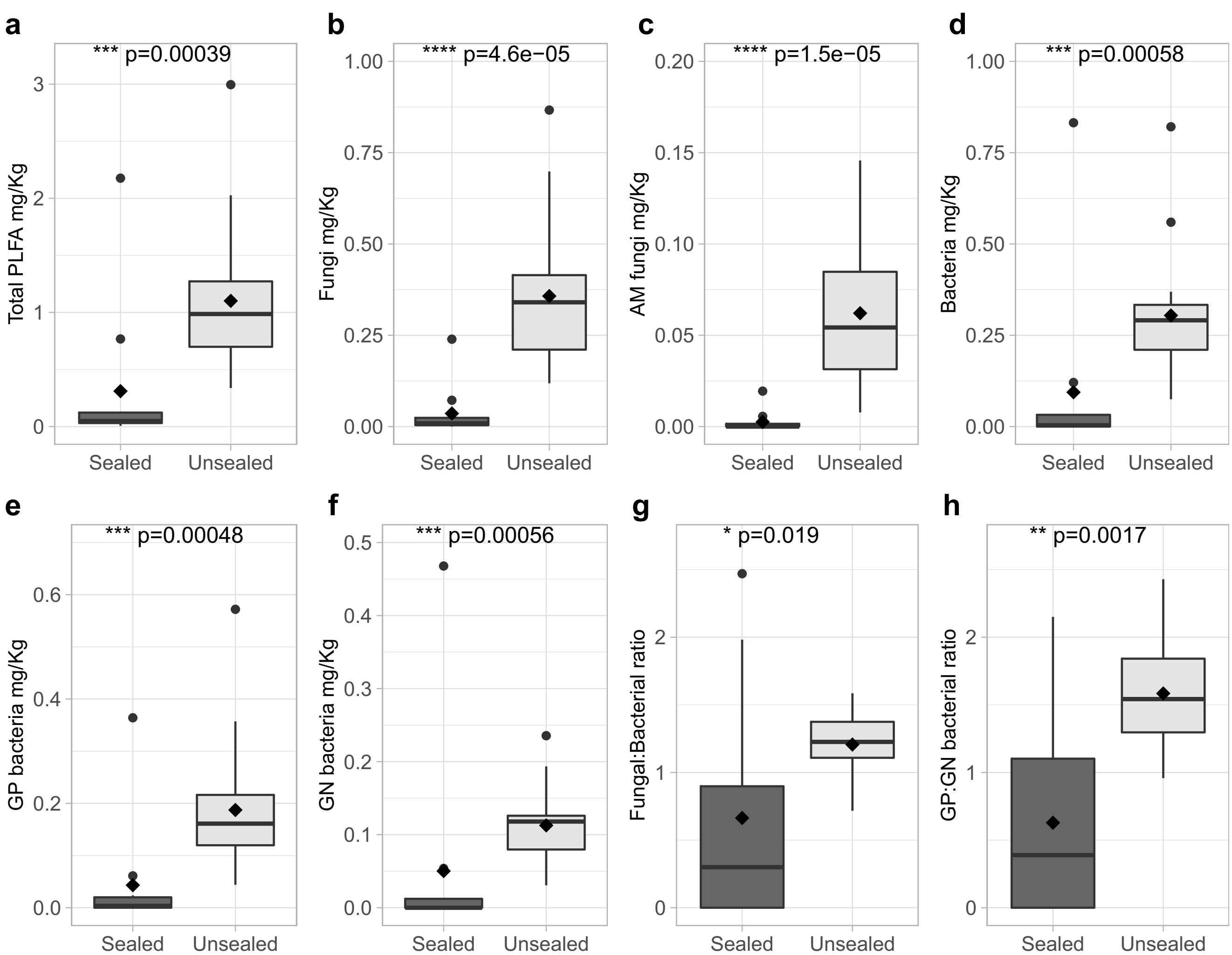

Fig. 4 Microbial community in sealed and unsealed soils. **a** Total PLFA, **b** total fungi, **c** AM fungi, **d** total bacteria, **e** GP bacteria, **f** GN bacteria, **g** fungal:bacteria ratio and **h** GP:GN bacterial ratio. A significant difference between sealed and unsealed soils was estimated by Wilcoxon test, with "****", "***" and "**" indicating significance at $p < 0.0001$, $p < 0.001$ and $p < 0.01$, respectively

Table 2 Spearman's rank correlation (rho) and *p* values of correlations between microbial attributes and soil properties in sealed and unsealed soils. Significant correlations with *p* values < 0.05 are indicated in italics

Microbial attribute	Soil status	Total C		Total N		C:N ratio		Water content	
		rho	*p* value	rho	*p* value	rho	*p* value	rho	*p* value
MBC	Sealed	0.31	0.356	0.61	0.052	−0.27	0.418	0.47	0.146
	Unsealed	0.50	0.051	0.20	0.450	0.35	0.188	0.41	0.114
N-mineralisation potential	Sealed	−0.04	0.902	−0.21	0.534	0.02	0.951	−0.18	0.598
	Unsealed	0.29	0.278	0.25	0.343	0.04	0.891	−0.04	0.891
Total PLFA	Sealed	0.57	0.071	0.55	0.082	0.13	0.714	0.55	0.087
	Unsealed	0.58	*0.020*	0.62	*0.012*	−0.08	0.771	0.75	*0.001*
Total fungi	Sealed	0.46	0.156	0.5	0.113	0.03	0.936	0.5	0.121
	Unsealed	0.59	*0.019*	0.54	*0.034*	−0.01	0.978	0.75	*0.001*
Total bacteria	Sealed	0.56	0.072	0.63	*0.038*	−0.13	0.696	0.71	*0.015*
	Unsealed	0.56	*0.025*	0.68	*0.005*	−0.13	0.633	0.68	*0.005*
Fungal:bacterial ratio	Sealed	0.23	0.499	0.09	0.802	0.19	0.574	0.23	0.499
	Unsealed	0.29	0.283	−0.22	0.404	0.48	0.064	0.03	0.926
GP bacteria	Sealed	0.56	0.072	0.63	*0.038*	−0.13	0.696	0.71	*0.015*
	Unsealed	0.52	*0.042*	0.69	*0.004*	−0.19	0.484	0.66	*0.007*
GN bacteria	Sealed	0.64	*0.032*	0.71	*0.015*	−0.19	0.569	0.79	*0.004*
	Unsealed	0.21	0.443	0.61	*0.015*	−0.42	0.104	0.65	*0.008*
GP:GN bacterial ratio	Sealed	0.42	0.203	0.55	0.079	−0.19	0.569	0.68	*0.022*
	Unsealed	0.52	*0.040*	0.47	0.070	0.01	0.969	0.33	0.217

properties were notably altered in sealed areas, with a reduction of total C, total N and water content, and a consequent increase in C:N ratio. Sealing had a negative impact on microbial attributes, with a large reduction of the microbial community (MBC and PLFA biomarkers) and activity (N-mineralisation). Additionally, microbial attributes that correlated with soil properties in unsealed soil did not show equivalent correlations in sealed soil, such as those between total PLFA and total fungi to total C, and total N and water content. These results suggest that the microbial community in sealed soil may respond differently to that in unsealed soil, indicating that sealing may disrupt the microbial response to changes in soil properties and lead to negative impacts on microbial services. The PLFA data provides an indicator of the microbial community in sealed soil, where low fungal:bacterial and gram-positive:gram-negative bacterial ratios indicate degradation in microbial C sequestration and a consequential effect on soil C storage in sealed soil.

4.1 Soil sealing leads to depletion of C, N and water content

The sealed soils exhibited lower total C, total N and water content than unsealed soils (Table 2 and Fig. 2a). Soil sealing leads to a reduction of soil C due to topsoil removal during the construction process and the reduction of C inputs from organic matter, plant root exudates and residue decomposition (Edmondson et al. 2012; Raciti et al. 2012; Wei et al. 2013, 2014; Piotrowska-Długosz and Charzyński 2015; Yan et al. 2015; Majidzadeh et al. 2017, 2018). Indeed, sealed soils have been recorded as having significantly lower C stores when compared with unsealed or greenspace soils in urban areas (Wei et al. 2014; Piotrowska-Długosz and Charzyński 2015; Majidzadeh et al. 2017). Additionally, if C decomposition continues within sealed soil, even at a low rate (Wei et al. 2014; Piotrowska-Długosz and Charzyński 2015), and there are negligible C inputs (Majidzadeh et al. 2018), this will contribute to C losses. In this context, elevation of microbial C respiration in sealed soil has been linked to increases in water content (Piotrowska-Długosz and Charzyński 2015; Majidzadeh et al. 2017, 2018). In sealed soil, water content is affected by the type and size of pavement or sealing surface (Morgenroth et al. 2013), and beneath impervious and semi-permeable pavements, the water content is, in general, lower than in greenspace soils (Hu et al. 2018; Piotrowska-Długosz and Charzyński 2015). In soil under semi-permeable surfaces, water moving from adjacent greenspaces into sealed soil can promote C inputs beneath sealed surfaces (Majidzadeh et al. 2018); however, this can also increase the microbial processes of C decomposition and lead to C losses (Majidzadeh et al. 2017, 2018). In soil under house crawl spaces of different ages, most C was lost in the first 50 years after construction, but after 50 years, C sequestration became the dominant process (Majidzadeh et al. 2018). Overall, it is not clear whether longer periods of sealing lead to an increase or decrease in the C balance of sealed soils, and this is an area which requires further investigation.

The notable depletion of total N, as seen in our results (Fig. 2b), is a commonly observed consequence of soil sealing, often being greater in magnitude than losses of total or organic C (Raciti et al. 2012; Zhao et al. 2012; Wei et al. 2014; Majidzadeh et al. 2018; Hu et al. 2018). Our results indicate that, in sealed soil, total N was reduced by over 60% compared with unsealed soil (Fig. 2b), whilst total C was reduced by nearly 40% compared with unsealed soil (Fig. 2a), leading to a higher C:N ratio in sealed soil (Fig. 2c). Our results are comparable to other observations of sealed soil where total C reduction was between 42 and 57%, and N depletion was between 47 and 97% (Majidzadeh et al. 2018; Piotrowska-Długosz and Charzyński 2015; Raciti et al. 2012; Zhao et al. 2012). The effect of sealing appears to be most notable and variable for N dynamics and processes which can be connected to the length of time sealed, organic C availability and water content, influencing the sealing impact on microbial processes (Zhao et al. 2012; Piotrowska-Długosz and Charzyński 2015; Majidzadeh et al. 2017, 2018) and N-mineralisation potential (Fig. 3b, Zhao et al. 2012). Previous research has shown that sufficient water content can promote microbial decomposition and N-mineralisation where there is available organic C (Zhao et al. 2012; Majidzadeh et al. 2018), leading to inorganic N production (Zhao et al. 2012; Majidzadeh et al. 2018), and potential leaching of NH_4^+-N and NO_3^--N and accumulation in the subsoil (Zhao et al. 2012). Where water can infiltrate into sealed soils from adjacent unsealed areas (Majidzadeh et al. 2018), we speculate that mineralisation of remaining organic matter could be stimulated. Considering, the reduced levels of C and the absence of plant roots, N assimilation by microorganisms and plants is likely to be low, resulting in N losses over time by leaching, subsoil accumulation and groundwater transport. Beyond that, these circumstances may lead to inorganic N pollution of urban groundwater and water courses (Zhao et al. 2012).

4.2 Sealing alters microbial attributes and community composition

Soil sealing leads to a drastic reduction in microbial attributes. Our results showed that sealed soil exhibited a reduction in MBC (Fig. 3a), as consistently reported in previous studies (Wei et al. 2013; Piotrowska-Długosz and Charzyński 2015; Majidzadeh et al. 2017, 2018). Observations of low MBC in sealed soil have commonly been associated with low C, N and water content (Wei et al. 2013; Piotrowska-Długosz and Charzyński 2015; Majidzadeh et al. 2017, 2018; Hu et al. 2018). Our PLFA data also demonstrated the negative impact of sealing on the microbial community (Fig. 3), with sealed soil exhibiting significantly lower mass of total PLFA and microbial markers, consistent with reductions in MBC, total C, total N and water content. It has been observed that a reduction in the microbial community reflects low microbial activity (Zhao et al. 2012; Piotrowska-Długosz and Charzyński 2015), a pattern also observed in our results with the significantly reduced N-mineralisation potential in sealed soil.

In studies of urban soil, few have considered the relationship between soil properties and microbial attributes in both sealed and unsealed soil. Indeed, physical and chemical properties, in particular water content, have been shown to have significant effects on microbial attributes in unsealed soils (Wei et al. 2014; Piotrowska-Długosz and Charzyński 2015) and have exhibited positive correlations with MBC, catalase activity and β-glucosidase activity in unsealed soil, but not in sealed soil (Piotrowska-Długosz and Charzyński 2015). Here, neither MBC nor N-mineralisation potential had significant correlations with any soil properties across sealed or unsealed soils. Conversely, the PLFA data does show significant responses of the microbial community to soil properties (Table 2). In unsealed soil, increases in C, N and water content correlated with growth of the microbial community (total PLFA, bacteria and fungi), which is typical for natural soils or those under agricultural conservation management (Helgason et al. 2014; Bai et al. 2020). However, in sealed soil, only bacteria were correlated with soil properties, suggesting that sealing disrupts the relationships normally seen in natural and agricultural soils between microbial attributes and soil properties. Positive correlations identified between both total N and water content (Table 2) could indicate that input of water and N promoted bacterial growth. Other studies have found additional soil properties associated with sealing-driven microbial depletion, including potassium and phosphorus availability, heavy metals and dissolved organic C (Hu et al. 2018; Yu et al. 2019). Low respiration and metabolic quotient observed on sealed soil (Piotrowska-Długosz and Charzyński 2015) can suggest organic matter of low quality. Thus, sealing results in alterations to soil properties and negative impacts on the soil microbial community and processes.

Sealing also caused alterations to the microbial community composition, notably the fungal:bacterial ratio and GP:GN ratio. The effect of sealing was seen more strongly in fungi, with sealed soils having ~ 93% less fungi than unsealed soils, and ~ 78% less bacteria than unsealed soils. Consequently, the fungal:bacterial ratio decreased in sealed soils indicating greater numbers of bacteria to fungi (Fig. 4g). Fungi have been shown to be resistant to conditions of low total N, high C:N ratio and low water content (Six et al. 2006; Strickland and Rousk 2010; Fang et al. 2020), conditions which are commonly observed in sealed soils. However, these conditions did not lead to greater dominance of fungi in this study. Conversely, soils affected by degradation processes such as tillage, deforestation, trampling and contamination usually present a greater impact on the fungal community and show a proportional decrease on the fungal:bacterial ratio (Kaur et al. 2005; Malmivaara-Lämsä et al. 2008; Simmons and Coleman 2008; Bischoff et al. 2016; Montiel-Rozas et al. 2018; Lopes and Fernandes 2020). Thus, our results suggest that fungi in sealed soils may be more affected by aspects of soil sealing not included in this study but that commonly arise due to the degradation processes of urbanisation, such as contamination and disturbance.

The decrease in the GP:GN bacterial ratio in sealed soil (Fig. 4h) suggests that GN bacteria are more adapted to sealing than GP bacteria. GN bacteria presented a positive correlation with total C, whilst GP bacteria had no correlation with total C (Table 2). As GN bacteria are more dependent on simple sugars (Kramer and Gleixner 2008; Fanin et al. 2019), the organic C that is promoting GN bacterial growth is likely to be labile and soluble C transported by water from adjacent greenspaces, a process which has been suggested as a source of organic C in soils beneath house crawl spaces (Majidzadeh et al. 2018). Additionally, GN and GP bacteria had positive correlations with total N and water content, suggesting there may also be transport of soluble N by water from adjacent greenspaces, and that this may be an important source of nutrients for bacteria in sealed soil.

In contrast to GN bacteria, GP bacteria are linked to more complex SOC (Kramer and Gleixner 2008; Fanin et al. 2019). Therefore, the low biomass of GP bacteria can be related to low levels of complex SOC remaining in sealed soil as a consequence of topsoil removal and microbial degradation over time.

4.3 Sealing limits the microbial community and affects the C storage service

Litter degradation plays an important role in C inputs into soil. Organic and inorganic compounds released during decomposition, and the remaining complex organic compounds are essential components of soil organic matter synthesis (Jastrow et al. 2007). In sealed soil, the sealed surface acts as a barrier preventing this source of organic C from reaching the soil, such that low or no organic C or nutrients from litter can enter the soil (Zhao et al. 2012; Majidzadeh et al. 2017, 2018), which in turn, affect soil biological and nutrient processes.

Plants and roots also contribute greatly to soil C stores. The lack of plants growing on sealed surfaces usually leads to a reduced root colonisation, limiting the C inputs from plant exudates and dead roots. Consequently, microbial processes that take place in the soil-root zone and depend on plant exudates are limited beneath sealed surfaces. Many of these processes are related to N inputs and nutrient availability, highlighting N biological fixation, N oxidation reactions and phosphate solubility (Sylvia et al. 2005; Paul 2007). Many fungal species establish a mutualistic association with plant roots to obtain organic molecules and, as payment, they colonise soil space to assimilate and transport nutrients directly back to the plant roots (Smith and Read 2008). By enhancing the soil microbial community, roots enable microbial processes connected with organic matter formation, such as the microbial release of biomolecules and dead biomass (Jastrow et al. 2007; Clemmensen et al. 2013). Thus, it is likely that the lack of plant and root growth, litter inputs and microbial activity in the soil-root zone all contribute to the lower C stores in sealed soil.

Fungal biomass in soil is, in general, suggested to contribute to high soil C storage (Strickland and Rousk 2010). Fungi exhibit low nutrient requirements and high C use efficiency which results in more C being allocated to their biomass, per unit of substrate used, compared with bacteria, which have lower C use efficiency (Six et al. 2006). Fungi have the ability to grow under a high C:N ratio, permitting their mycelial growth to explore wider areas and translocate nutrients across the soil (Strickland and Rousk 2010). In addition, fungal biomass is more complex and resistant to decomposition than bacterial biomass, introducing a more stable form of organic C in the soil (Jastrow et al. 2007; Clemmensen et al. 2013). Whilst studies have presented different insights into the functional implications of the fungal:bacterial ratio (Strickland and Rousk 2010; Soares and Rousk 2019), in general, a higher fungal:bacterial ratio is assumed to promote an increase in soil organic matter (Jastrow et al. 2007; Strickland and Rousk 2010). Therefore, the observed reduction in fungi and consequent bacterial dominance in sealed soil is likely to lead to notable limitations to C storage.

The lower GP:GN bacteria ratio in sealed soil illustrates that there is more GN bacteria to GP. This indicates that there is less recalcitrant C in the sealed soil (Kramer and Gleixner 2008; Fanin et al. 2019), which suggests the reduced ability of sealed soils not only to store C but to store it as stable C that may be more protected from decomposition (Lal 2004; Marschner et al. 2008), highlighting the wider impacts of soil sealing on the ecosystem service of soil C storage.

5 Conclusion

Soil properties were notably affected in sealed soil, with a large significant reduction in total C, total N and water content in sealed soils. Microbial biomass C, N-mineralisation potential and microbial PLFA markers were also significantly reduced in sealed soils. Our results show that changes to soil properties, caused by sealing, led to a drastic decrease in the microbial community and important microbial processes. The increase of the C:N ratio and decrease of the F:B and GP:GN ratios suggest that sealed soils are degraded due to the loss of C, which limits fungal and bacterial growth. In addition, the reduced inputs of C from litter degradation and plant exudates, associated with the reduction of fungal dominance, indicate a limitation on the C storage potential of sealed soil. Furthermore, the correlation of bacteria with C, N and water suggests that there may transport of soluble C and N by water into sealed soils from adjacent greenspaces. This may be an important source of nutrients for microbes in sealed soil, and the investigation of this process would be beneficial to further understand sealed soil nutrient cycling and implications for C and N fluxes. In this context, further work, such chronosequence studies, would elucidate how urbanisation and soil sealing impact the dynamics of C and N and microbial processes over time, and as a consequence, the ecosystem services of sealed soil.

Authors contribution All authors contributed to the study conception and design. Material preparation, data collection and analysis were performed by Carly Stevens and Marlon Correa Pereira. The final draft of the manuscript was written by authors Marlon Correa Pereira and Roisin O'Riordan, and all authors contributed to previous versions of the manuscript. All authors read and approved the final manuscript.

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