The influence of impactor size cut-off shift caused by hygroscopic growth on particulate matter loading and composition measurements

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Highlights:

- Hygroscopic growth leads to a shift in the size of dry particles cut off by impactors used in measurements of particle mass and composition.
- We propose a method for evaluating this influence on analysis of aerosol composition, quantifying its global importance for the first time.
- Observational comparisons and model validation must account for the large temporal and spatial variations in this influence.

Abstract:

The mass loading and composition of atmospheric particles are important in determining their climate and health effects, and are typically measured by filter sampling. However, particle sampling under ambient conditions can lead to a shift in the size cut-off threshold induced by hygroscopic growth, and the influence of this on measurement of particle loading and composition has not been adequately quantified. Here, we propose a method to assess this influence based on κ-Köhler theory. A global perspective is presented based on previously reported annual climatological values of hygroscopic properties, meteorological parameters and particle volume size distributions. Measurements at background sites in Europe may be more greatly influenced by the cut-off shift than those from other continents, with a median influence of 10-20% on the total mass of sampled particles. However, the influence is generally much smaller (<7%) at urban sites, and is negligible for dust and particles in the Arctic. Sea-salt particles experience the largest influence (median value ~50%), resulting from their large size, high hygroscopicity and the high relative humidity (RH) in marine air-masses. We estimate a difference of ~30% in this influence of sea-salt particle sampling between relatively dry (RH=60%) and humid (RH=90%) conditions. Given the variation in the cut-off shift in different locations and at different times, a consistent consideration of this influence using the approach we introduce here is critical for observational studies of the long-term and spatial distribution of particle loading and composition, and crucial for robust validation of aerosol modules in modelling studies.

Keywords: aerosol measurement; chemical composition; filter sampling; growth factor.

Graphical abstract:

1. Introduction

Particulate matter (PM) is currently of major concern around the world due to its effects on human health (Meister et al., 2012; Pope et al., 2009) and climate change (IPCC, 2013; Ramanathan & Carmichael, 2008; Seinfeld & Pandis, 2006). The mass loading and composition of particles are crucial factors in these effects (Putaud et al., 2004; Seinfeld & Pandis, 2006; Van Dingenen et al., 2004). Filter sampling and subsequent laboratory analysis is one of the major ways to obtain particle composition and loading (Chow, 1995; Mader & Pankow, 2001; Schaap et al., 2011). An impactor (or inlet head) is employed at the start of the airflow to cut off particles larger than a certain size (Aufschnaiter, 2009; Berner & Luerzer, 1980; Marple et al., 1991; Schauer et al., 2003), so that PM_1, $PM_{2.5}$ and PM_{10} (particulate matter with an aerodynamic diameter smaller than 1, 2.5 and 10 μm, respectively) can be collected on filters for offline analysis. However, ambient particle diameter is strongly influenced by aerosol liquid water (ALW), a ubiquitous and abundant aerosol constituent (Heintzenberg 1989; EPA 1999), whose amount is governed by particle hygroscopic growth. Global ALW may exceed dry particle mass by a factor of more than two (Lee & Adams, 2010; Liao & Seinfeld, 2005; Nguyen et al., 2016). Therefore, particle hygroscopic growth and ALW of ambient particles can significantly influence the cut-off diameter of dry particles, and hence lead to a considerable shift in the cut-off that impacts the particle composition and loading measured using filter-based sampling.

The cut-off shift is determined by particle hygroscopicity, of which large variations have been observed on a global scale (Liu et al., 2011; Nguyen et al., 2016; Pringle et al., 2010; Wu et al., 2013a, 2013b; Wu et al., 2011). Furthermore, hygroscopic growth is strongly influenced by relative humidity (RH), which also varies greatly in space and time (Köppen, 1900; Dai, 2006; Willett et al., 2014). Measurements made in different seasons and locations may suffer the cut-off shift to different extents. Without sufficient understanding of the cut-off shift, it is difficult to derive consistent seasonal variations and yearly trends of particle composition or compare different locations. Moreover, insufficient knowledge of the cut-off shift can hinder effective model validation. Most modelling results provide the mass loading and chemical composition of dry particles (e.g., Archer-Nicholls et al., 2015; Mann et al., 2014; Mann et al., 2010; Tsyro, 2005; Zaveri et al., 2008), but filter measurements report results under ambient RH (i.e., wet particles). The impact of this on model validation is expected to be important over humid

regions, e.g. marine and coastal regions where high RH and abundant hygroscopic sea-salt particles are both present (Chen et al., 2016; Neumann et al., 2016a, 2016b).

Despite the important influence of hygroscopic growth and cut-off shift on particle composition and loading measurements, there is no efficient way to dry the airflow for a high-volume aerosol sampler. The airflow better not be heated or dried during the sampling, since these processes change the gas-particle equilibrium and lead to a loss of semi-volatile compounds (Chow, 1995; Grassian, 2001; Mader & Pankow, 2001; Schaap et al., 2011; Van Dingenen et al., 2004; Shingler, et al., 2016;), e.g. nitrate (Chow et al., 2005; Hering & Cass, 1999; Schaap et al., 2004; Slanina et al., 2001; Vecchi et al., 2009; Chen et al., 2018) and secondary organic aerosols (Iinuma et al., 2010; Mader & Pankow, 2000). As recommended by EPA and WMO/GAW (EPA, 1999; WMO/GAW, 2016), the inlet should sample the particles under ambient conditions. It is worth noting that some experiments dry airflow in inlet manifolds due to different scientific focuses, this would lead to evaporative losses of semi-volatile compounds and Shingler et al. (2016) estimated the losses based on thermo-kinetic simulations. Consequently, the influence due to cut-off shift is inherent in measurements of particle composition, and requires full investigation and careful assessment to ensure consistent comparisons between measurements.

In this study, we propose a method to assess the influence of this cut-off shift on particle composition and loading measured using filter-based sampling. A global perspective on the influence is presented first, based on κ-Köhler theory (which describes the hygroscopic growth of particles, Köhler, 1936; Petters & Kreidenweis, 2007) together with previously reported annual climatological values of hygroscopic properties, meteorological parameters (RH and temperature) and particle volume size distributions. This work enables us to quantify the influence of cut-off shift on analysis of particulate matter loading and composition, provides a firm foundation for more consistent studies of the long-term characteristics and spatial distributions, and brings more confidence to model validation.

2. Method and Data

To explore the influence of cut-off shift on filter-based particle sampling driven by hygroscopic growth, we calculate the aerodynamic diameter of ambient particles based on the hygroscopic diameter growth factor (GF). The GFs are derived from κ-Köhler theory (Petters & Kreidenweis, 2007), with consideration of particle composition and size as well as

meteorological conditions (Eq. 1). In this study, particles are assumed to be homogeneously internally mixed with the same chemical constitution throughout each size mode, and the influence of the cut-off shift on total particulate matter loading is assessed. The influence on each particle component can also be assessed, although there is uncertainty resulting from the assumption about the particle mixing state (discussed in the section 3.3). More precise assessment of the influence of cut-off shift on particle components can be derived using the approach we propose here if detailed observations of particle mixing state and size-segregated composition are available. The collection efficiency, which depends on aerodynamic design of the impactor, air flow rate and pressure drop (Hillamo & Kauppinen, 1991; Marple et al., 1991; Wang & John, 1988), influences the sampled particles. This will influence the cut-off shift, which is generally less than 10% and is discussed in detail in section 3.4. In this study, we assume that the influence of sampling collection efficiency is corrected during chemical post-processing, so that particles larger than the nominal cut-off thresholds are totally blocked by the impactor and are not collected on the filter.

The κ-Köhler theory proposes a single parameter (κ) to describe the hygroscopic properties of particles, representing the dependence of hygroscopicity on chemical composition. The original Köhler equation (Köhler, 1936) can be transformed into the following Eq. (1), which describes the relationship between RH, κ and dry particle diameter (Liu et al., 2014; Petters & Kreidenweis, 2007):

$$RH/100 = \frac{GF^3 - 1}{GF^3 - 1 + \kappa} \exp\left(\frac{4\sigma_{s/a} M_w}{R(T + 273.15)\rho_w D_{dry} GF}\right) \quad (1)$$

where *RH* has units of [%], *GF* is the diameter of an ambient particle divided by its dry diameter (D_{dry}, [m]), *T* is the temperature [°C], $\sigma_{s/a}$ is the surface tension of the solution–air interface, *R* is the universal gas constant, M_w is the water molar weight, ρ_w is the water density, and κ is the hygroscopicity parameter. More details of the κ value and density of each components from previous studies are given in Table S1, please see also the references (Asa-Awuku et al., 2010; Bond & Bergstrom, 2006; Cross et al., 2007; Fountoukis & Nenes, 2007; Liu et al., 2014; McMeeking et al., 2010; Petters & Kreidenweis, 2007; Pringle et al., 2010).

The κ value of an internally mixed particle can be derived from its composition using the Zdanovskii-Stokes-Robinson (ZSR) mixing rule (Stokes & Robinson, 1966; Zdanovskii, 1948), which weights the κ of each component according to its respective volume fraction. A previous

modelling study (Pringle et al., 2010) provided the global distribution of κ for fine (0.1-1 μm) particles, showing a fair agreement with most measurement-derived κ. The deviations between the modelled and observed κ values were less than 0.05 at 10 out of the 14 locations (Pringle et al., 2010). We adopt their statistical κ value for each continent in this study (Tables 1 and 2 in Pringle et al., 2010), and assume that the coarse particles (1-10 μm) have the same κ as fine particles in the absence of size-resolved κ studies. The measured chemical properties or size-resolved κ of particles is applied where available (Marine, Beijing and Melpitz cases described below). O'Dowd et al. (2004) reported that coarse marine particles are very close to pure sea-salt with more than 90% sodium chloride, whereas fine marine particles consist of more organic components. Therefore, κ of sodium chloride is adopted for coarse marine particles in this study; while the κ of fine marine particles follows Pringle et al. (2010). Liu et al. (2014) investigated the size-resolved κ based on chemical composition measurements at a background station on the North China Plain (NCP) near Beijing. The HOPE-Melpitz campaign (part of the HD(CP)2 Observational Prototype Experiment, Macke et al., 2017) provided size-segregated (PM_1 and PM_{10}) chemical ions and composition from filter measurements with a high-volume DIGITEL DHA-80 sampler (Walter Riemer Messtechnik, Germany), more details of sampling and laboratory analysis are given elsewhere (Spindler et al., 2013; Spindler et al., 2004). The ISORROPIA II thermodynamic model (Fountoukis & Nenes, 2007) is used to derive the inorganic components (e.g. ammonium nitrate and ammonium sulfate, please see details in Table S1 and Fountoukis & Nenes, 2007) from ions measured by filter sampling at Melpitz. We then follow the approach proposed by Liu et al. (2014) to derive κ for fine and coarse modes based on their chemical composition. Homogeneous mixing is assumed for all chemical components in fine and coarse modes in the absence of size-resolved particle composition information. Therefore, κ is different between fine and coarse modes, but is consistent within each mode.

Dry particle volume size distributions (PVSD) can be described by a multimodal distribution, namely a lognormal distribution in the fine/accumulation (0.1-1 μm) and coarse (> 1μm) modes (Whitby, 1978). In this study, we follow the typical PVSDs in different air-mass/aerosol-type/site categories in Whitby (1978), which are based on a compilation of global in-situ measurements. For a specific case at Melpitz, we derive the PVSD from measurements using an Aerodynamic Particle Sizer (APS Model 3320, TSI, Inc., Shoreview, MN USA) and a Twin Differential Mobility Particle Sizer (TDMPS, Birmili et al., 1999, TROPOS, Leipzig, Germany). Both APS and TDMPS were operated under dry conditions, and measured particle

number size distribution in the coarse and fine modes, respectively. PVSD is then calculated assuming a spherical shape. The mobility diameter is converted to an aerodynamic diameter following Chen, et al. (2016) and Heintzenberg et al. (1998). It is worth noting that fine particles also include the Aitken mode (< 0.1 μm), which is not considered in this study since it is negligible for the mass/volume loading and too small to be cut off.

To estimate the hygroscopic growth and cut-off shift, we use the annual climatological global distribution of surface RH reported by (Dai, 2006) for each continent and for marine regions. This is based on an analysis of long-term (1976-2002) global measurements from over 15,000 weather stations and ships. A recent study (Willett et al., 2014) based on 1976-2005 global measurements reported annual climatological surface temperatures (T) and RH, and shows a good agreement with Dai (2006). The reported surface temperature is adopted for each continent, and a fixed surface temperature of 26.85^{o}C (300 K) is assumed over marine regions due to absence of data. Although this does not provide a realistic T variation, GF is insensitive to T in the range -30 ^{o}C to +30 ^{o}C, and less than 5% difference is observed for a 1 μm sea-salt particle when RH=90%, according to κ-Köhler theory (Fig. S1). For specific cases in Beijing (Liu et al., 2014) and Melpitz (September 2013), the average RH and T observed during the corresponding sampling periods were used.

3. Results and Discussion

The cut-off shift due to aerosol hygroscopic growth is ubiquitous in particle composition measurements made with filter-based sampling. Fig. 1 and Table S2 summarize the influences of cut-off shift on particle sampling (PM_1, $PM_{2.5}$ and PM_{10}) for different site/aerosol-type categories following the classification in Whitby (1978), including marine surface (sea-salt), desert dust, tropical forest, urban-average, average background, clean continental background, background and aged urban plume, as well as urban and freeway. These assessments are based on annual climatological datasets, but it is worth noting that the cut-off shift can vary substantially during different measurement periods. The resulting influence for a specific observation can be assessed using the corresponding measurement data via this proposed approach, as shown later for specific measurement campaigns in Beijing and Melpitz.

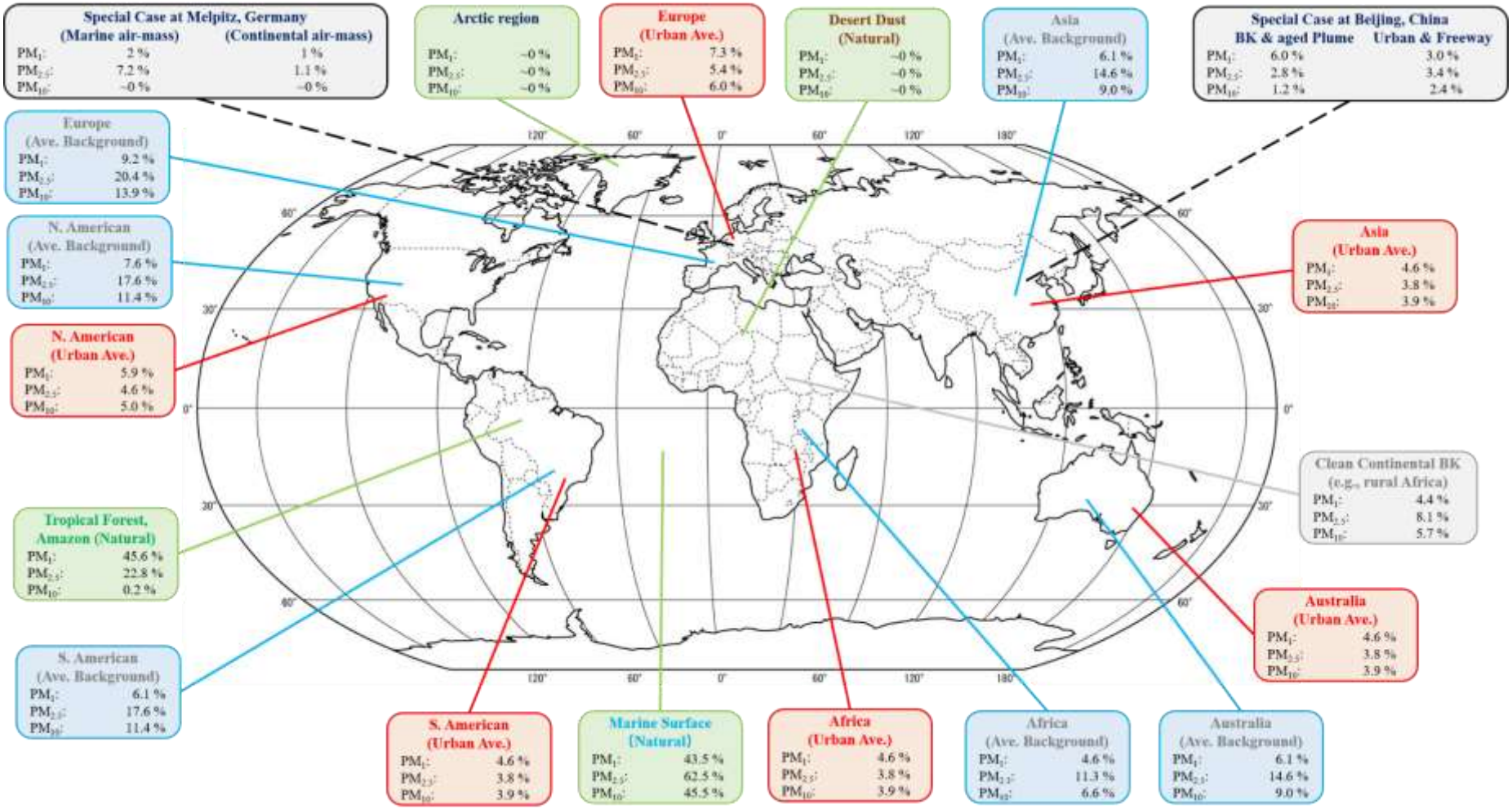

Figure 1. The influence of cut-off shift on particle mass loading. Green box (natural source aerosols), red box (urban-average over continents), blue box (average background over continents), grey box (clean continental background, taking Africa for example), and black box (two specific cases: Beijing & Melpitz). Annual climatological values are shown in this figure, more detailed parameters and uncertainties are given in Table S2 and S3.

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3.1 Influences of the cut-off shift on particle measurements

Of particles from natural sources, sea-salt experiences the most significant influence of cut-off shift, due to its high hygroscopicity (Twomey, 1954; Petters & Kreidenweis, 2007), high volume/mass fraction at large sizes (Fig. S2), and high RH over marine surfaces (Dai, 2006). As shown in Fig. 2, the influence on sampled sea-salt particles in $PM_{2.5}$ can exceed 80% when RH is higher than 90%. Over marine surfaces, as much as ~60% (median value) of sea-salt is estimated to be additionally cut off by a $PM_{2.5}$ impactor/inlet head compared with dry conditions, and up to ~45% by a PM_1/PM_{10} impactor (Table S2). In contrast, there is almost no influence on hydrophobic dust particles (Pringle et al., 2010) and Arctic particles. Arctic particles rarely reach sizes larger than 0.5 μm (Heintzenberg & Leck, 2012; Heintzenberg et al., 2006; Korhonen et al., 2008), far below the cut off threshold. The influence on PM_{10} sampling is negligible for biogenic particles over tropical regions, due to their low hygroscopicity ($\kappa \sim = 0.1$, (Pringle et al., 2010) and very limited volume/mass fraction around 10 μm (Huffman et al., 2012). However, the influence on tropical forest particles sampled by PM_1 impactor can reach up to ~45%, and more than 20% by $PM_{2.5}$ impactor. This is because there is a large contribution from biogenic particles with sizes around 1 μm and 2.5 μm to the total volume of PM_1 and $PM_{2.5}$, respectively (Huffman et al., 2012), see Fig. S2. Generally, the influence of cut-off shift on filter-based sampling is negligible for dust, but should be comprehensively considered for aerosols over marine and coastal regions, as well as tropical forest. Greater care should be taken for a size-segregated particle composition analysis, where the cut-off shift not only influences the total PM mass, but may also shift particles from smaller size bins to larger bins.

Over regions influenced by anthropogenic activities, our estimated ALW is consistent with previously reported values (Nguyen et al., 2016) all over the world. The highest contribution of ALW (in percentage) to continental ambient total particle mass concentrations is found to be in Europe. ALW contributes 22-56% (median 40%) of ambient total particle mass concentrations in an urban-average air-mass over Europe (Table S2), but less than 30% over other continents. This is because urban-average particles generally have a relatively low hygroscopicity, with median κ values less than 0.3 on continents other than Europe, where a higher median κ value of 0.36 (Pringle et al., 2010) and a relatively humid atmosphere (average RH~=75%, Dai, 2006) are both present. In an extreme case with high RH and highly hygroscopic particles present simultaneously, the contribution of ALW to the ambient total

particle mass concentrations may reach up to ~95% in a European average background air-mass, much higher than in an urban-average air-mass. In general, average background particles are larger than urban-average particles (Whitby, 1978), resulting in a reduction of the Kelvin effect and therefore an enhancement in water vapour condensation and cut-off shift. Correspondingly, the median value of the resulting influences on PM_1/$PM_{2.5}$/PM_{10} sampling over European cities (urban-average sites) can be 5.5-7.5%, and may reach 50-60% (median value ~10-20%) for average background sites. Over other continents, the influence for urban sites is almost negligible (<5%), and for average background sites is about 10-17% for $PM_{2.5}$ and less than 10% for PM_1 and PM_{10}. Taking rural regions in Africa as an example of clean continental background, the influence is ~5% for PM_1 and PM_{10} measurements and ~8% for $PM_{2.5}$ measurements. Reported measurements over the NCP/Beijing region in summer 2007 are adopted in this study as examples of 'background and aged urban plume' and 'urban and freeway' (Liu et al., 2014), and the influences are found to be negligible (<6%) for PM_1, $PM_{2.5}$, and PM_{10} here.

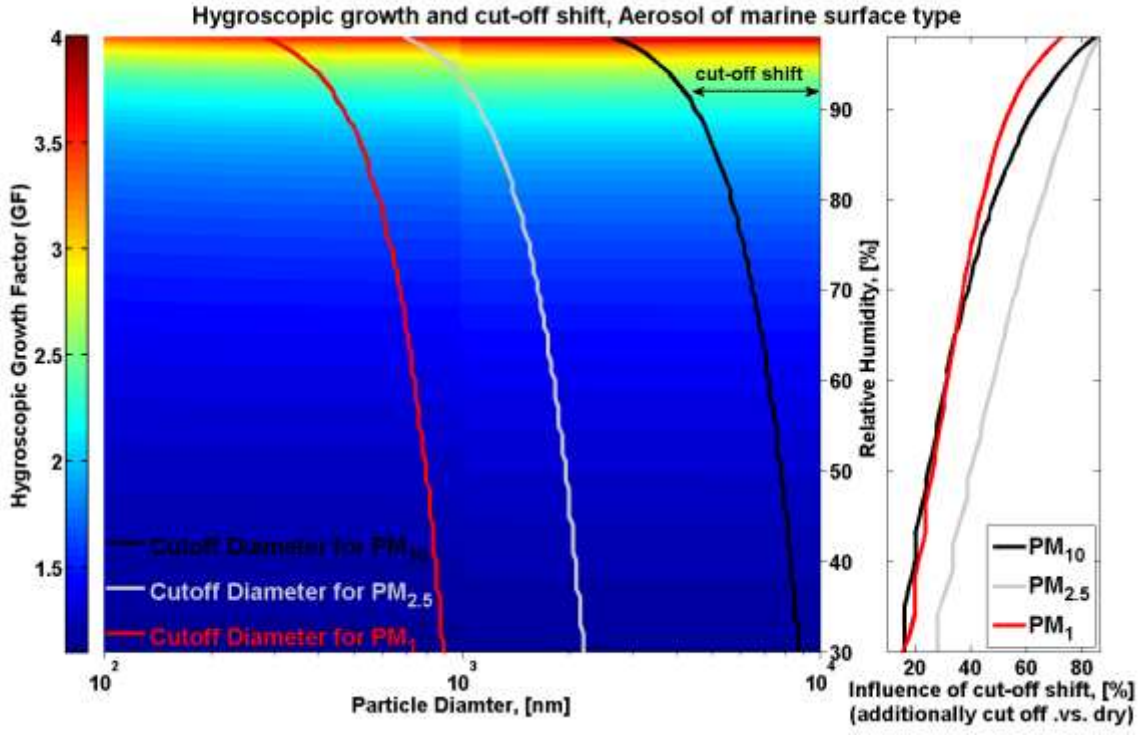

Figure 2. The hygroscopic diameter growth factor (GF, left panel) of marine surface aerosol, including fine mode (considerable organic components) and coarse mode (nearly pure sea salt), under different RH condition. The cut-off point of dry particles under different RH condition is indicated by the colourful lines (red: PM_1; grey: $PM_{2.5}$; black: PM_{10}). The influences of cut-off shift on particle volume concentrations are given in the right panel.

3.2 Variability of the cut-off shift

The influence of the cut-off shift varies greatly with time as well as location. Understanding this variability is critical for study of the long-term and spatial distribution of particle composition and for model validation. Observational data from the HOPE-Melpitz campaign are used to illustrate this variability. Generally, the influence is small (<10%) at Melpitz, a central European background site, where the coarse particle volume concentration is very limited during this campaign (Fig. S3). However, the influence rapidly increased from ~1% to ~7% for $PM_{2.5}$ measurements (Fig. 1) when a marine air-mass brought hygroscopic coarse particles to Melpitz (Fig. S3 and Fig. S4). Super-micron (1-2.5 μm) sea-salt particles experienced substantial hygroscopic growth, and were strongly influenced by the cut-off shift. This issue is of great importance for measurements at marine and coastal locations, where sea-salt is an important contributor to total particulate matter. As shown in Fig. 2, there is a 30% difference in the sampled sea-salt mass between RH=60% and RH=90% conditions due to the cut-off shift alone.

3.3 Impact of mixing state and aging process on cut-off shift

The mixing state of particles can impact the cut-off shift. For instance, aging processes can mix sea-salt with sulfate and organic carbon (OC) internally via heterogeneous reaction and condensation, moderating the hygroscopic growth of sea-salt particle and influencing the cut-off shift during sampling. Taking this aging process of sea-salt as an example, we conduct a simplified theoretical investigation of the impact of mixing state on the cut-off shift. We make the simplified assumptions that sea-salt is homogeneously internally mixed with more hygroscopic sulfate (Na_2SO_4 with κ=0.76, Fountoukis & Nenes, 2007; Liu et al., 2014) and less hygroscopic water soluble organic carbon (OC with κ=0.3, Asa-Awuku et al., 2011; Asa-Awuku et al., 2010). A consistent percentage of sulfate and OC is assumed throughout all particle sizes, and this percentage is used to indicate the extent of aging. We use the PVSD for marine aerosol shown in Table S2, and assume that it remains unchanged during the aging process. The Zdanovskii-Stokes-Robinson (ZSR) mixing rule is applied to calculate the κ values of aged sea-salt-sulfate and sea-salt-OC particles, respectively. As shown in Fig. 3, the influence of cut-off shift on the measured sea-salt mass loading decreases as aging increases. This reduction in the influence is larger for $PM_{10}/PM_{2.5}$ than PM_1 for sea-salt, and can decrease

by ~10% for sea-salt-sulfate and by 15-20% for sea-salt-OC particle once the aging level reaches 70%. The influence of cut-off shift reduces more slowly as RH falls. The influence of

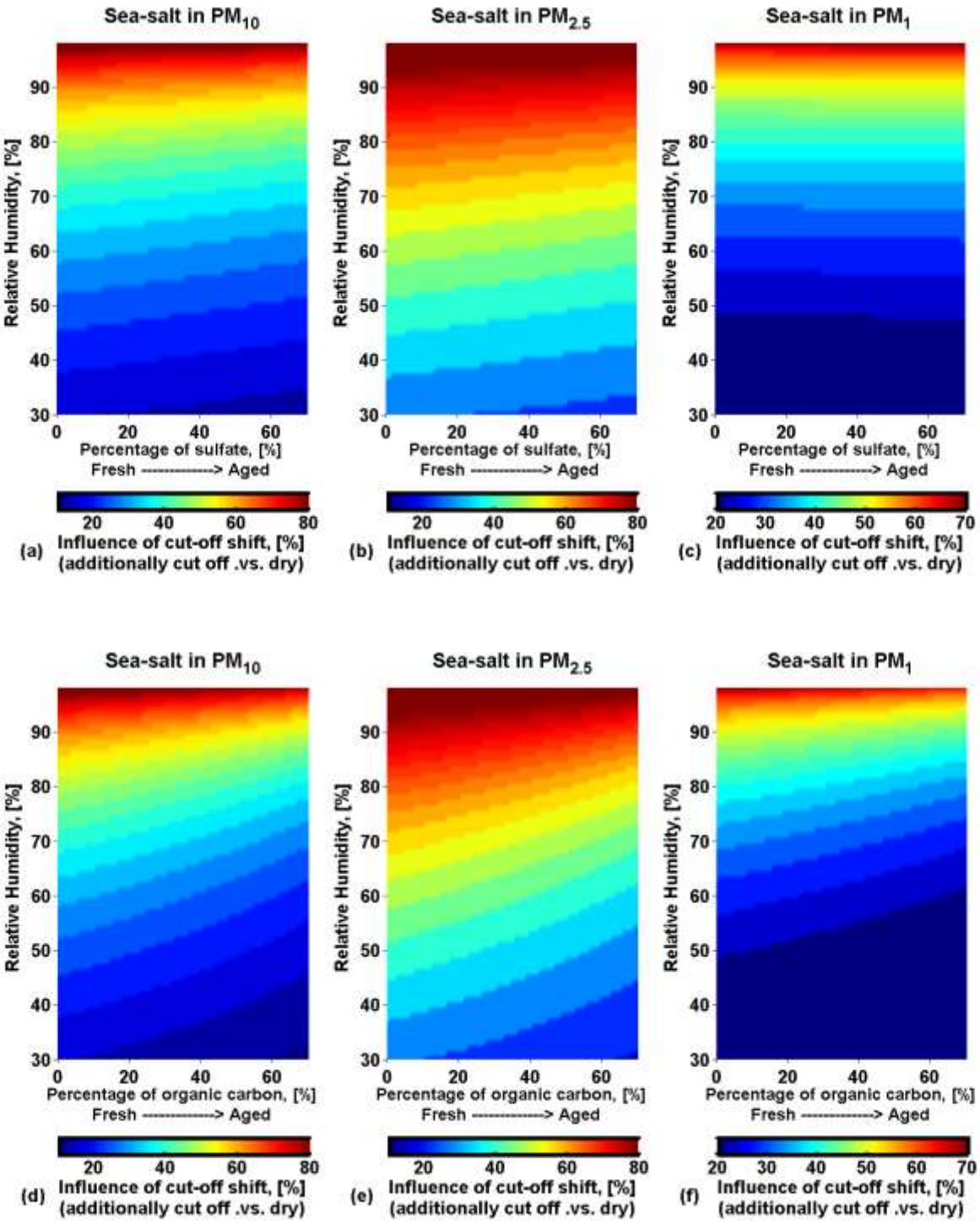

Figure 3. Impact of mixing state or aging process on the cut-off shift. **(a, b, c)** internally mixed between sea-salt and sulfate; **(d, e, f)** internally mixed between sea-salt and water soluble organic carbon. The percentage of sulfate or organic carbon indicates the aging level of sea-salt particles.

cut-off shift for sea-salt-OC particles is more sensitive to the aging level than sea-salt-sulfate particles, due to the different hygroscopicity of sulfate and OC. Conversely, mixing with sea-salt enhances the cut-off shift in measurements of sulfate and OC. For example, with RH=90%, the influence of cut-off shift on sulfate measurements increases from 60% to 70% in PM_{10} (70% to 80% in $PM_{2.5}$) when the percentage of sea-salt increases from 30% to 80% (Fig. 3a and 3b). Similarly, with RH=90%, the influence of cut-off shift on OC measurements increases from 55% to 70% in PM_{10} (65% to 80% in $PM_{2.5}$) when the percentage of sea-salt increases from 30% to 80% (Fig. 3d and 3e). Detailed observations of mixing state are therefore needed in future field measurements to refine assessment of the influence of cut-off shift on each particle component.

3.4 Impact of collection efficiency on cut-off shift

The collection efficiency, depending on aerodynamic design of the impactor, air flow rate and pressure drop influences the particle sampling. A ~2% over-sampling of sea-salt due to the shallow collection efficiency curve (Fig. 4) is found, and this may decrease the influence of cut-off shift. Here, we take the sampling of sea-salt particles with two PM_{10} impactors as examples to illustrate the impact of collection efficiency on cut-off shift. The PVSD of sea-salt (Whitby, 1978) and collection efficiency curves of demonstrated impactors, TEOM PM_{10} low volume sampler (EPA, 1991) and impactor (Marple et al., 1991), are shown in Fig. 4. Taking the collection efficiency curve into account, the influence of cut-off shift decreases slightly. Generally, this impact of collection efficiency on cut-off shift increases slowly as RH increases; however, as shown in Fig. 4, it remains less than 10% for the demonstrated shallow efficiency curve (EPA, 1991) and less than 5% for the steep curve (Marple et al., 1991). This impact of collection efficiency on cut-off shift is expected to be smaller on the $PM_1/PM_{2.5}$ sampling of sea-salt or sampling other types of aerosol. Since a single peak close to the cut-off threshold is essential for a sensible impact of efficiency curve on particle sampling or cut-off shift. Dust has a similar single peak PVSD to sea-salt, but it is hydrophobic and therefore there is no cut-off shift due to hygroscopic growth.

3.5 Impact of particle volume size distribution on the cut-off shift

To investigate the impact of PVSD on the cut-off shift, we conduct sensitivity studies by varying the geometric mean diameter by volume (DGV), standard deviation (σ) and volume

concentration (V) of PVSD by ±10%, as shown in Table S3. Two types of aerosol are chosen as examples. The first is marine surface aerosol, representing highly hygroscopic aerosol predominantly in the coarse mode; and the second is averaged urban aerosol in Asia, representing less hygroscopic aerosol with clearly defined fine and coarse modes in PVSD. Negligible impact (<2%) on the cut-off shift is observed for the second aerosol type due to its limited hygroscopicity. No impact from V can be observed for either aerosol type, since varying V does not change the shape of PVSD. Small differences (1-3%) in the influence of cut-off shift can be observed due to the variation of DGV in both aerosol types, although this effect could be larger in aerosols with a DGV lying close to the cut-off threshold of the impactor. However, a large impact can be observed for the highly hygroscopic marine aerosol with a ±10% variation of σ. Although the differences for $PM_{2.5}$ and PM_{10} marine aerosol are generally smaller than 5%, an 8-14% difference in the influence of cut-off shift can be observed for PM_1 marine aerosol when RH=60% (and 5-11% when RH=90%). The particle size distribution is important for assessing the cut-off shift, and therefore it is very helpful if this can be measured during field campaigns.

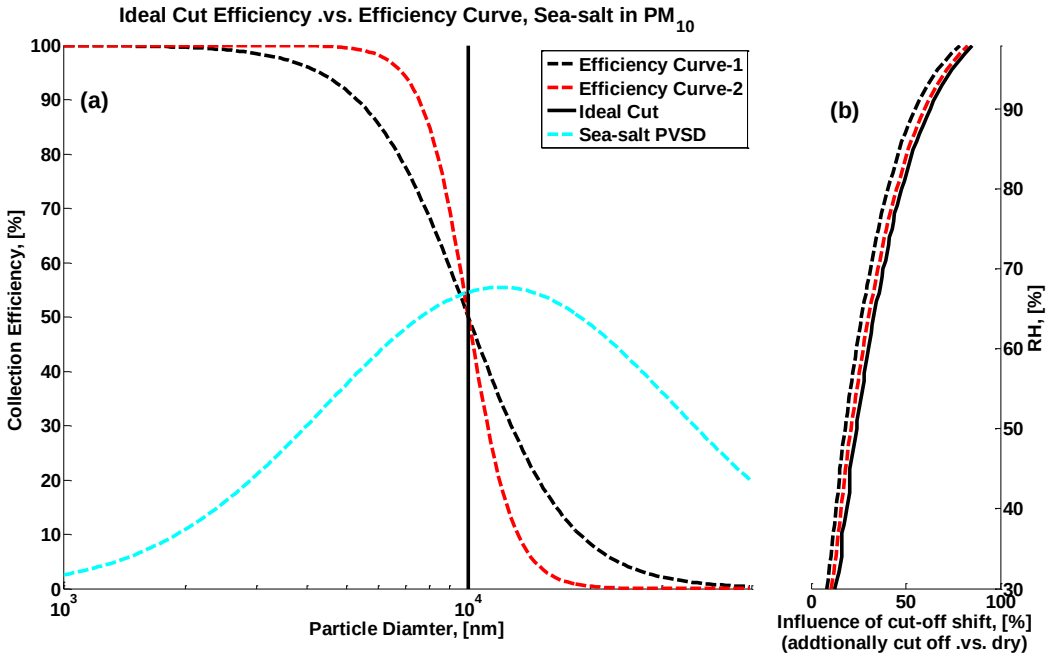

Figure 4. Impact of collection efficiency on cut-off shift during sea-salt sampling. Left panel **(a)** shows the PVSD (light blue dashed line) of sea-salt, an ideal efficiency curve (black solid line) and two collection efficiency curves of demonstrated impactors. The dashed black line indicates the TEOM PM_{10} low volume sampler (shallow curve, EPA, 1991); and the dashed red line indicates a steep-curve impactor (Marple et al., 1991). Right panel **(b)** shows the corresponding influences of

cut-off shift with consideration of two collection efficiency curves (dashed lines) and ideal cut (black solid line).

4. Conclusions

This study highlights the importance of the shift in the size of dry particles cut off by impactors, resulting from hygroscopic growth. A method for assessing the influence of this cut-off shift on analysis of particulate matter loading and composition is proposed, and an algorithm in MATLAB language is provided in the supplementary materials. We present the first global perspective of this influence.

Large temporal and spatial variation of the influence is found. On average, particles over marine surfaces are found to experience the largest influence of cut-off shift, where as much as ~60% of particle mass is estimated to be additionally cut off by a $PM_{2.5}$ impactor compared with sampling under dry condition, and up to ~45% by a PM_1/PM_{10} impactor. In contrast, the cut-off shift has no influence for dust and particles in the Arctic region. The influence for biogenic particles in tropical forest is negligible for PM_{10} measurements, but considerable (23-46%) for $PM_{2.5}$ and PM_1 measurements. The influence is generally negligible (less than 7%) over urban areas, but need to be considered (about 10-20%) over continental background areas. This influence needs to be assessed for each measurement period individually even at the same location, since it is highly dependent on the ambient conditions. We estimate a difference of ~30% in this influence of sea-salt particle sampling between relatively dry (RH=60%) and humid (RH=90%) conditions. Our sensitivity studies show that detailed measurements of particle size distribution and mixing state are helpful for refining assessments of this influence.

This work proposes a method to quantify the influence of cut-off shift on analysis of particulate matter loading and composition, and to investigate the variability of this influence from a temporal and spatial perspective. It is critical for observational studies focusing on long-term and spatial distribution of particle loading and composition, and crucial for robust validation of aerosol modules in modelling studies.

Supplementary Materials

Figure S1-S4;

Table S1-S3;

The MATLAB script in the '.m' files.

Acknowledgments

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