Opinion: how will advances in aerosol science inform our understanding of the health impacts of outdoor particulate pollution?

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Abstract. Air pollution, with high levels of particulate matter (PM), poses the greatest environmental threat to human health, causing an estimated seven million deaths annually and incurring 5% of the global GDP. While PM health impacts are influenced by the toxicity of its individual chemical constituents, the PM mortality burden is solely based on its total mass concentration. This is because of a lack of large-scale, high-resolution PM chemical composition data needed for epidemiological assessments. Identifying which PM constituents are harmful for health has been the 'Holy Grail' of atmospheric science, since the seminal six US cities study that first linked PM to mortality in 1993. Ever since, atmospheric scientists have focused on understanding aerosol composition, emission sources and formation pathways, while longitudinal epidemiological studies needed individual level exposure data, using land use regression models for the prediction of exposures at fine resolutions. In this opinion article, we argue that the time has now come to shift focus towards considering PM chemical composition in epidemiological health assessments, laying the foundation for the development of new regulatory metrics. This shift will enable targeted guidelines and subsequent regulations, prioritizing mitigation efforts against the most harmful anthropogenic emissions. Central to this shift is the availability of global long-term, high resolution PM chemical composition data obtained through field observations and modelling outputs. In the article, we underscore key milestones within aerosol science integral for advancing this foundational shift. Specifically, we examine emerging modelling tools for estimating exposure to individual PM components, present the type of ambient observations needed for model developments, identify key gaps in our fundamental understanding of emissions and their atmospheric transformation and propose a forward cross-disciplinary collaboration between aerosol scientists and epidemiologists to understand the health impacts of individual PM components. We contend that aerosol science has now reached a pivotal moment in elucidating the differential health impacts of PM components, as a first step toward their incorporation into air quality guidelines.

1. Preamble

1.1 A brief chronology of air pollution

A tale of global air pollution has already been narrated by Fowler et al., and only a brief chronology will follow, presenting the main milestones reached by the atmospheric science community since the earliest recorded accounts of air pollution (Fowler et al., 2020). The threat of air pollution to human health has been recognized since the time of Hippocrates, around 400 BCE (Jones et al., 1923). Successive written accounts of air pollution occur throughout the following two millennia until

measurements from the eighteenth century onwards demonstrated the growing scale of poor air quality in urban centres. One of the most emblematic early historical documents on air pollution was published in 1661 under the title *Fumifugium* by Evelyn (Evelyn, 1772). Evelyn documented air pollution in London and proposed solutions for reducing the scale of the problem by moving industries to the countryside. Graunt, a contemporary of Evelyn, observed a correlation between rates of mortality and pollution, especially during fog episodes, albeit in the absence of any chemical data or numerical values to quantify the pollutants present (Graunt, 1939). Later, in 1775, Sir Percival Pott was one of the first to document the effects of specific pollutants on health. Pott observed a high incidence of scrotal cancer among chimneysweepers and concluded that exposure to soot was a risk factor for the cancer (Brown and Thornton, 1957).

The industrial revolution accelerated the growth and geographical spread of emissions, as highly polluted cities became the defining problem that culminated with the great smog of London in 1952. This pollution episode of a few days duration caused an estimated 10,000 deaths and injury to more than 100,000 persons (Stone, 2002; Bell et al., 2004). The London smog is believed to be the worst air pollution event in the history of the United Kingdom and the most notorious for its effects on environmental research, government regulation, and public awareness of the relationship between air quality and health. It was instrumental for establishing an unambiguous link between short-term exposure to peak levels of pollution and acute health effects. It also led to the introduction of the Clean Air Act of 1956 that aimed to reduce emissions and mitigate future pollution events. Until the latter decades of the twentieth century, Europe and North America dominated global emissions and suffered the majority of adverse health and environmental effects. By that time, the transboundary issues of acid rain (Egnér and Eriksson, 1955) and ground-level ozone (Volz and Kley, 1988; Fowler et al., 2008) were the focal environmental and political air quality problems (Vasseur, 1973). As emission controls took effect in the Western world, Asia rapidly industrialized, emerging as the primary contributor to global emissions by the early twenty-first century.

Towards the end of the 20th century, the health effects of air pollution resurfaced as a top priority, as new epidemiological evidence highlighted this time the breadth of chronic health problems resulting from long-term pollution exposures (Brunekreef and Holgate, 2002). For this, the emergence of extensive networks of surface measurements, satellite remote sensing, and numerical models was indispensable for providing global air quality data with which epidemiologists could estimate the adverse health effects of air pollution. Since then, numerous studies have documented the chronic and acute health effects of air pollution and burden of disease, many with a global perspective (Burnett et al., 2018; Cohen et al., 2017; Mcduffie et al., 2021; Richard T. Burnett, 2014; Lelieveld et al., 2015; Lelieveld et al., 2019; Chen et al., 2018b; Chen and Hoek, 2020; De Bont et al., 2022; Holtjer et al., 2023; Nyadanu et al., 2022). Today, air pollution remains a major public health concern, and efforts continue to reduce emissions and improve air quality.

1.2 Particulate air pollution

The polluted air we breathe can contain high levels of particulate matter, PM, commonly termed aerosols. PM is a complex mixture of tiny solid or liquid particles suspended in the air, with a size ranging from a few nanometers to a few micrometers (John H. Seinfeld, 2016). These particles can be directly emitted from primary sources, or be formed in the atmosphere by gas-to-particle conversion of secondary oxidation products, e.g. sulfate from SO_2 oxidation, nitrate from NO_X oxidation or secondary organic aerosol (SOA) from the oxidation of volatile organic vapours. Primary PM sources can be either natural or human-made. Natural sources include desert dust, sea-spray, wildfires and biogenic SOA

from the oxidation of plant volatiles, while anthropogenic sources include emissions from residential heating or car exhaust and their secondary oxidation products. As a result, PM has an immensely complex chemical composition with different levels of toxicity depending on the emission sources and/or formation processes (Hallquist et al., 2009; Jimenez et al., 2009). Smaller particles are more likely to enter our bloodstream and travel deep into our lungs, causing damage. Short-term exposure to peak levels of PM can cause acute health effects (Dockery et al., 1993; Liu et al., 2023; Nhung et al., 2017). By contrast, long-term exposure can lead to chronic diseases, such as cardiovascular, cerebrovascular (De Bont et al., 2022) and respiratory diseases (Holtjer et al., 2023), which are responsible for a large share of the air-pollution-related mortality (Burnett et al., 2018; Cohen et al., 2017; Chen and Hoek, 2020). Current epidemiological evidence reveals that no level of air pollution can be deemed safe, and even low levels of PM may carry significant risks (Strak et al., 2021; Pinault et al., 2016; Cohen et al., 2017; Dominici et al., 2022; Brunekreef, 2021; Brauer et al., 2019). Today, according to the Global Burden of Disease project, PM pollution is responsible for an estimated seven million deaths every year (Burnett et al., 2018). This positions PM among the five leading causes of deaths worldwide, alongside high blood pressure, smoking, diabetes and obesity (Cohen et al., 2017).

1.3 PM mitigation: a global challenge of the 21st century

Although particles are compositionally heterogeneous with notable temporal and spatial variations, most studies investigating their adverse health effects used total PM mass concentration, treating them as a uniform entity. This is because total PM mass concentration was routinely measured, primarily as $PM_{2.5}$ in the USA and PM_{10} in Europe[1], and used as the basis of exposure assessment – either directly or modelled – in epidemiological studies. As a result, and based on the accumulated evidence, PM mass serves today as the primary metric for particulate pollution regulation.

In 2021, the World Health Organization, WHO, updated its air quality guidelines to propose a much more stringent limit value for $PM_{2.5}$ of 5 $\mu g\ m^{-3}$ (Who). These new guidelines provide a basis to justify aggressive regulation of anthropogenic emissions to improve global air quality. However, such low $PM_{2.5}$ concentrations are currently only found in few remote environments, and over 95% of the world population lives in places where the new guidelines are not met. Several western countries have made significant progress over the past 20 years to meet the former WHO limit for $PM_{2.5}$ of 10 $\mu g\ m^{-3}$ last updated in 2005 (Southerland et al., 2022; Hammer et al., 2020). In contrast, $PM_{2.5}$ levels exceeding 50 $\mu g\ m^{-3}$ are typical in low- to middle-income countries, e.g. in Eastern-Europe, China or India, where 90% of attributable deaths occur (Lelieveld et al., 2015). This translates to a loss of several years of life expectancy in Asia due to pollution, compared to several months in the West (Lelieveld et al., 2019).

Reducing fossil fuel dependence and residential emissions will undoubtedly significantly improve air quality, especially in polluted environments (Pai et al., 2022; Mcduffie et al., 2021). However, natural sources including desert dust, wildfires and biogenic emissions alone will impede many regions from complying with the new WHO guidelines. This, however, is not a reason to dismiss the evidence-based limit values set by the WHO in 2021 that are set to protect health, but to improve on them. A recent landmark modelling analysis suggests that over 50% of the global population will still be living in places with $PM_{2.5}$ concentrations greater than 5 $\mu g\ m^{-3}$, even if all anthropogenic emissions would be eliminated (Pai et al., 2022). Moreover, natural emissions are likely to increase in the near future due to climate change, further complicating efforts to meet the 2021 WHO guidelines in certain regions (Gomez et al., 2023). Meeting these guidelines will be particularly challenging for many

[1] $PM_{2.5}$ and PM_{10}: Particulate matter with a size lower than 2.5 and 10 μm, respectively.

regions worldwide, and globally applicable solutions to manage and improve air quality will become no longer evident. This entails a complete rethink of how we should be mitigating air pollution and underlines a need for a new generation of air quality metrics that focus on specific anthropogenic PM components in addition to total PM mass.

Targeting particulate pollution across individual chemical components allows for the consideration of their varying degrees of toxicity, a concept known as the "differential toxicity of PM components" (Masselot et al., 2022). PM health effects are mediated by their size, solubility, and chemical composition, and hence their sources and formation processes. In our recent work, we have identified the organic and metal fractions to be of particular concern for oxidative stress (Daellenbach et al., 2020) and inflammation (Leni et al., 2020), in contrast to secondary inorganic particles that dominate PM mass. Given that oxidative stress and inflammation are one of the pathway to chronic disease development (Mudway et al., 2020), this necessitates a closer inspection of which sources of PM should be mitigated. It is therefore vitally important for aerosol scientists to provide global PM chemical composition data, enabling epidemiologists to pinpoint the most detrimental components and empowering policymakers to implement targeted, cost-effective measures to reduce specific health-relevant anthropogenic PM sources across different regions.

1.4 Introductory overview

In this account, we discuss how the broader atmospheric science community can inform policies and interventions to mitigate sources of PM components that pose risks to human health (Figure 1). We advocate for a foundational shift towards considering PM deferential toxicity in epidemiological health assessments, made possible through improved air quality modelling suitable for exposure assessment, and present the key milestones within aerosol science that, in our view, are necessary for this shift. Section 2 introduces the concept of PM differential toxicity and its potential as an exposure metric. Section 3 critically examines recent advances in modelling tools for estimating fine-scale exposures to specific PM components. In section 4, we identify the type of ambient observations we think are essential for developing and validating exposure models. In section 5, we highlight remaining gaps in our understanding of PM component emissions, their atmospheric transformation and associated health effects and identify research opportunities. Section 6 delves into the need for strong collaboration between research communities to elucidate biological mechanisms underlying the health impacts of specific PM components.

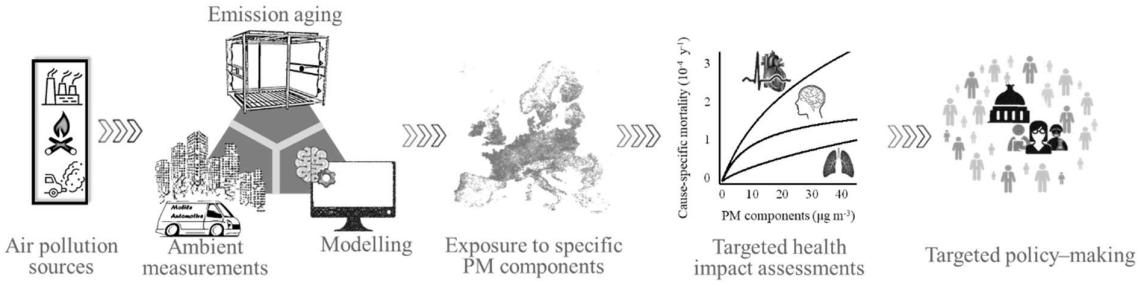


Figure 1: The multidisciplinary framework needed to target PM differential toxicity in policies.

2. Integrating PM chemical composition in health studies

2.1 PM differential toxicity as targeted air quality metric: more than just PM mass

To quantify the health impacts of PM, we currently rely on dose-response relationships, ideally derived from individual-level data from large cohort studies, that link morbidity and mortality to the

concentration of total PM mass. Whilst these relationships are from a broad evidence base, there is significant heterogeneity in the estimated effect size. This variation can be partially attributed to imperfect models approximating exposures or random differences among study populations. Yet, perhaps a large source of error lies in relying solely on PM mass concentration, ignoring the biological activity of different particle constituents. Although some studies have attempted to examine the adverse health outcomes of individual PM components, particularly highlighting associations with combustion and road traffic emissions, such investigations remain relatively infrequent due to the paucity of exposure data. As a result, PM chemical composition has not yet been included into any regulatory framework.

With the advent of vast amounts of atmospheric data, the time has now come to redirect our focus towards elucidating dose-response relationships for individual PM constituents. In practical terms, these constituents must be quantifiable, easily accessible, and readily available at the high spatio-temporal resolutions needed for exposure assessment, and across large spatial scales. Our proposal includes considering the following constituents: organic aerosol, elemental carbon, sulfate, nitrate, ammonium, sea-salt, brake-wear, and dust (Table 1). While brake-wear and dust concentrations cannot be directly measured, they can be traced using specific markers, such as Cu for brake-wear and Al for dust. The organic fraction should be ideally subdivided based on a source sectors, including primary and secondary aerosols from car exhaust, residential burning, wildfires, and biogenic emissions. Organic aerosol classes also cannot be directly measured, though might be retrieved through receptor modelling based on spectrometric measurements or chemical transport modelling, as discussed below. The classification of aerosols based on their chemical composition will also establish a direct link to aerosol sources (Table 1), offering policy makers effective and operational strategies for selectively mitigating the most important PM sources for health.

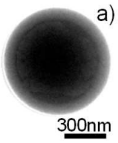
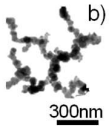
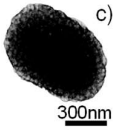
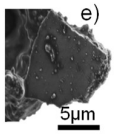
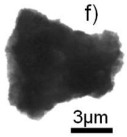
Beyond PM chemical composition, various other properties have been proposed to mediate aerosol health effects, including aerosol size, number, solubility, and oxidative potential (Table 1). For example, toxic metals primarily induce oxidative damage in their soluble form (Fang et al., 2017; Wong et al., 2020), whereas insoluble particles like asbestos or elemental carbon are biopersistent in the body, leading to chronic inflammation. Likewise, small particles can penetrate deep into the lungs, traverse the bloodstream and breach the blood-brain barrier, playing a role in respiratory, cardiovascular and neurological diseases (Requia et al., 2017; Maher et al., 2016), while a notable portion of large particles may be ingested leading to an imbalance in our gut microbiome (Fouladi et al., 2020; Alderete et al., 2018; Bailey et al., 2020). Parameters for emerging metrics intended to be used in future epidemiological studies should be standardized and widely available, which is currently unachievable for some of these parameters. PM chemical composition, which is now widely available, is intertwined with these alternative metrics (Table 1) and can constitute an effective mean to address PM health effects.

2.2 Necessity of PM chemical composition data at fine resolutions

More than 70% of the world population is projected to live in urban areas by 2050, with these densely populated agglomerations already being the most polluted environments (Mcduffie et al., 2021). The composition and concentrations of PM in these areas exhibit significant spatial heterogeneity from street to citywide scales. In some cases, intra-city variability exceeds the variability between different cities (De Hoogh et al., 2016; Eeftens et al., 2016; Tsai et al., 2015; De Hoogh et al., 2013; Eeftens et al., 2012a; Zhang et al., 2015; Jedynska et al., 2015). Such spatial heterogeneity is driven by traffic patterns (Simon et al., 2017; Li et al., 2016a; Gu et al., 2018; Elser et al., 2018; Elser et al., 2016),

restaurant emissions (Gu et al., 2018), domestic heating emissions (Elser et al., 2018; Elser et al., 2016; Jedynska et al., 2015; Mohr et al., 2011), industrial point sources (Shairsingh et al., 2018) and local geography (Mohr et al., 2011). Atmospheric aging of urban emissions and long-range transport of polluted air masses adds to this complexity, affecting PM background levels and composition on regional scales.

Table 1: PM chemical components, reflecting differential toxicity, suggested to be monitored and modelled to support health research. Components' physical properties that are important determinant of health effects are shown, including size, morphology, and solubility. Components' major sources are also shown. a), b), and c) Transmission electron microscopic images of organic aerosol, elemental carbon aggregates and sulfate adapted from (Li et al., 2011). d) Scanning electron microscopic image of a fresh sea-salt particle adapted from (Li et al., 2016b). e) Scanning electron microscopic image of a coarse brake ware particle adapted from (Kukutschová and Filip, 2018). f) Transmission electron microscopic image of a mineral dust particle adapted from (Xu et al., 2021).

Component	Size	Morphology	Solubility	Source
Organic aerosol	Fine		Moderately soluble for POA Soluble for SOA	Natural and anthropogenic, primary and secondary
Elemental carbon	Fine		Insoluble	Biomass and fossil fuel combustion
Sulfate	Fine		Soluble	Aqueous (65%) and gas phase OH (35%) oxidation of SO_2 from natural marine emissions (15%) and anthropogenic emissions from electricity generation and industries (85%) (John H. Seinfeld, 2016)
Nitrate	Fine		Soluble	Oxidation of NO_X emissions mainly from traffic exhaust
Ammonium	Fine		Soluble	Condensation of gas-phase ammonia mainly from agriculture emissions producing ammonium sulfate and nitrate
Sea salt	Coarse		Soluble	Natural marine emissions through bursting bubbles at the air-sea interface
Brake wear (Cu)	Coarse		Depending on the element	Brake pads
Mineral dust (Al, Si, Ti, Fe)	Coarse		Depending on the element and atmospheric age	Natural wind-blown desert dust on a global scale, in addition, to resuspended dust on an urban scales

Urban microenvironments strongly affect long-term exposures to specific PM components (Figure 2A). For example, studies have shown a link between road proximity, exposure to ultrafine

particles, and respiratory, cardiovascular, and neurodegenerative diseases (Alexeeff et al., 2018; Bayer-Oglesby et al., 2006; Yuchi et al., 2020; Boogaard et al., 2022). It has also been shown that exposures to high particle concentrations around train stations during typical daily commutes of less than one hour can contribute up to 21% of total daily PM exposure and more than 50% of daily exposure to key toxic metals such as Cu (Van Ryswyk et al., 2017). Given the substantial spatiotemporal variability in both PM concentrations and composition, it is imperative to determine PM chemical composition at fine spatial and temporal scales relevant to daily human activities. This understanding is vital for evaluating exposures to specific PM components and ultimately their health effects.

In epidemiological analyses, outdoor PM concentrations at residences are commonly used as proxies for exposure. While there is evidence supporting this approach, its applicability across different settings requires further investigation (Wei et al., 2023). As we spend most of our time indoors and new buildings are increasingly airtight for energy saving, outdoor concentrations may not reflect indoor levels (Schweizer et al., 2007). While indoor emissions, primarily from cooking (Klein et al., 2019) and smoking (Hyland et al., 2008), may influence health, they represent a separate risk factor distinct from outdoor air pollution, akin to contaminated water. This is because (1) different regulatory frameworks are needed to address emissions from indoor and outdoor sources, (2) these sources are often distinct, and (3) they require different control measures. Unlike outdoor air pollution, which often requires collective and regulatory abatement strategies to control emissions, indoor air pollution can be more effectively managed at the individual or household level, by improving ventilation and eliminating or reducing indoor sources. In the absence of indoor emissions, indoor concentrations are 30 to 70% lower than outdoors (Chen and Zhao, 2011) due to variability in infiltration rates. Moreover, exposures can also be influenced by outdoor pollution in other settings, such as workplaces and during commuting, where we spend a large fraction of our time. Health data from citizen cohorts often include questionnaires that offer valuable insights into indoor infiltration rates, workplace conditions and individual's mobility. While we consider outdoor concentrations at residence to be a reasonable proxy of exposure to outdoor pollution, integrating such information can help refining exposure estimations. First, however, the issue of downscaling air quality models to finer resolutions must be tackled.

3. Modelling exposures to individual PM components

The investigation of acute health effects requires the time-series analysis of daily exposures to specific components, typically obtained at an urban background site through long-term measurements (> 3 years) or modelling outputs. By contrast, longitudinal epidemiological studies of chronic diseases require long-term exposures determined at high spatial resolution – ideally at address level. Because high resolution PM composition data are scarce, existing epidemiological analyses considering PM chemical composition have mainly focused on acute effects, while health effects resulting from chronic exposure to individual PM components have rarely been assessed. In this section, we define the state-of-the-art in modelling PM exposure and discuss how recent advances in modelling PM chemical composition can help informing our understanding of PM differential toxicity.

3.1 Existing modelling approaches

Figure 2B compares three traditional classes of approaches used for estimating PM exposures. We put forward eight criteria for comparing these approaches including accuracy, spatial and temporal resolution, spatial and temporal coverage, capability of hindcasting and forecasting required to estimate past and future exposures and finally, source-specificity and chemical complexity, i.e. capability to quantify specific PM components.

Early cohort studies used averaged (Pope Iii et al., 2002) or interpolated (Jerrett et al., 2005) PM concentrations measured at routine monitoring stations to characterize the exposure of individual participants in different cities. However, stationary PM measurements are spatially sparse and do not account for the heterogeneity in pollutant concentrations within cities, especially for primary combustion emissions (Eeftens et al., 2012b; Elser et al., 2016; Elser et al., 2018). Therefore, several geo-statistical and process-based chemical transport models (CTMs) have been proposed to fill spatial gaps.

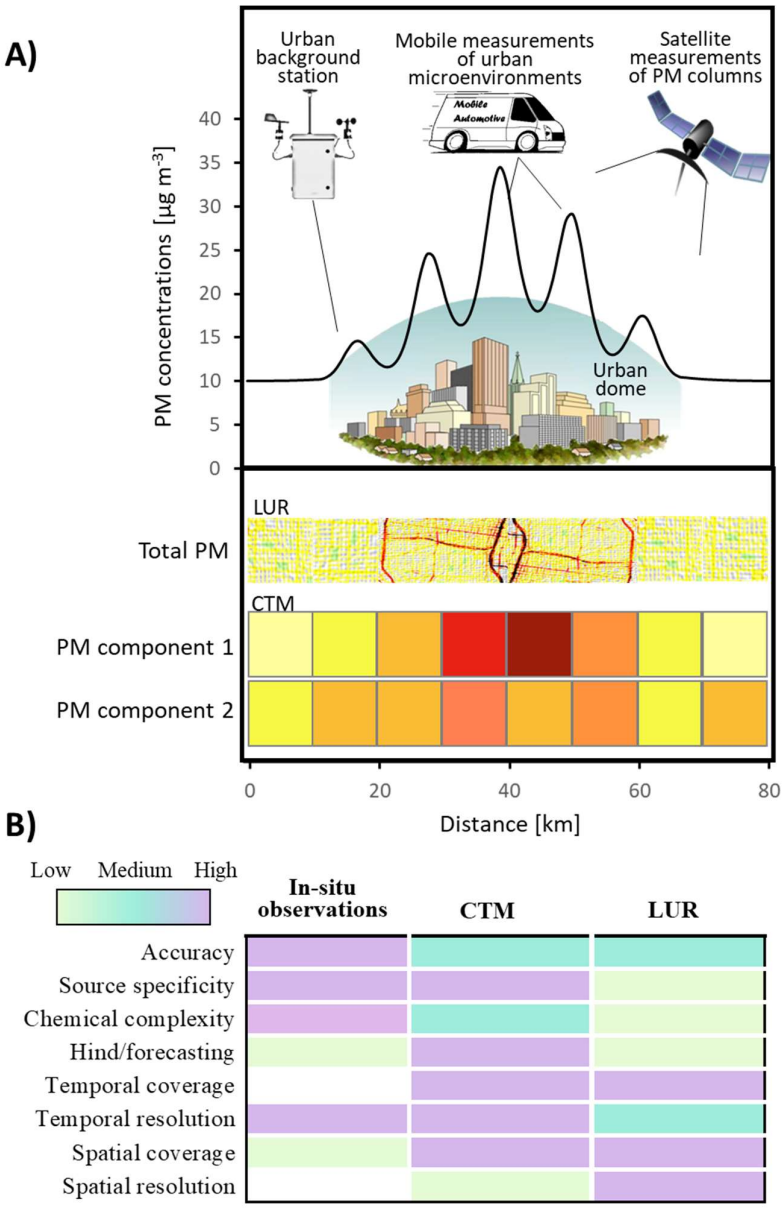


Figure 2: A) Representation of urban PM pollution, highlighting the urban increments in PM concentrations over background levels and the presence of microenvironments. State-of-the-art measurement and modelling strategies of PM concentrations at different scales are presented and compared in **B)** in terms of their advantages and limitations. Three different approaches are compared including field observations, chemical transport modelling (CTM) and land-use regression models LUR). The temporal coverage and spatial resolution of in-situ observations are determined by the method employed to obtain them, with white cells being assigned accordingly. Comparison of the performance of CTM vs. LUR is illustrated in **A)**, showing the source specificity of CTM and the high resolution of the LUR.

Land-use regression (LUR) models combine in situ observations with GIS based data, e.g. land use, traffic, or population density, as emission indicators to predict ground level PM concentrations on fine grids using regression techniques (Cattani et al., 2017; De Hoogh et al., 2016; De Hoogh et al., 2013; Eeftens et al., 2016; Hoek et al., 2011; Kim et al., 2016; Wolf et al., 2017). These techniques are covered in a review by (Hoek, 2017). LUR techniques are especially pertinent for modelling primary PM components, e.g. metals (Kim et al., 2016; Chen et al., 2020b) or combustion products (Jedynska et al., 2014; Jedynska et al., 2015) in addition to PM mass (Eeftens et al., 2012a). Typically, these LUR models predict annual means, suitable for longitudinal cohort studies on chronic health effects. Their accuracy is moderate, influenced by the number of monitoring sites (Wang et al., 2013) and the inclusion of source-specific predictor variables.

With advances in satellite remote sensing, aerosol optical depth, AOD, measurements of entire atmospheric columns have been introduced for assessing exposure to ground level total PM mass with much higher accuracy and relatively high time-resolution. Because AOD-PM relationships are non-linear, interactive, and spatiotemporally variable, AOD measurements are typically combined with other predictors including land-use data and meteorological variables. Machine-learning-based models using geo-statistical and AOD data have been successfully applied at different scales, including city, regional, national and continental scales as well as in different areas around the world, including EU, US, and China (Brokamp et al., 2017; Suleiman et al., 2016; Huang et al., 2018; De Hoogh et al., 2018; Di et al., 2016; Hu et al., 2017; Paciorek et al., 2008; Strawa et al., 2013; Zhan et al., 2017; Di et al., 2019; Xue et al., 2019; Chen et al., 2018b). However, because they are based on past AOD measurements, these models cannot forecast future PM concentrations, e.g., as a response to specific mitigation strategies (Figure 2B). More importantly, they are typically not capable of discriminating between specific PM components, because AOD measurements of PM columns are not yet chemically resolved, although future satellite-based sensors will partially deliver this capability (David et al., 2018).

Unlike the other methods, CTMs possess the ability to generate spatial and temporal distributions of chemically resolved PM components and forecast their future evolutions over large spatial scales. CTMs are bottom-up, process-based, numerical models, which simulate PM primary emissions and secondary formation, along with their losses and atmospheric transport in large 3-D Eulerian gridded domains. Despite their spatial coverage, source-specificity, and capability to leverage complex atmospheric oxidation processes, most CTMs are not sufficiently spatially resolved to be suitable for exposure assessments (Figure 2B). Due to computational constraints, highly resolved CTMs are currently limited to city scales. As a result, until very recently, CTM outputs have rarely been exploited for epidemiological analysis, except as an input variables for optimizing the retrieval of total PM mass concentrations in hybrid LUR models (Di et al., 2019; Xue et al., 2019; De Hoogh et al., 2016; Shen et al., 2022).

The two fields of air quality modelling, specifically using LUR and CTMs, have evolved along separate trajectories over the past three decades. This separation can be attributed to the modest accuracy of CTMs decades ago and their coarse spatial resolution, struggling to represent the substantial contribution of local pollution. Epidemiologists have then applied LUR techniques taking advantage of their high spatial resolution, reliance on locally measured air pollution data, ease of application, minimal input data requirements and moderate-to-good performance (Hoek, 2017). At that time, CTM developments have primarily focused on implementing representative emission and chemical schemes, aiming to enhance their accuracy. However, with the advancement in CTMs and the increasing regional nature of PM pollution, it is now the time for these two fields to converge to achieve accurate estimation

of exposure to individual PM components at high temporal and spatial resolution and coverage, fulfilling all eight criteria described in Figure 2B.

3.2 How recent advances in modelling PM chemical composition can help informing our understanding of PM differential toxicity?

More recent modelling developments have allowed the production of fine-resolution maps of PM chemical constituents on continental and global scales (Van Donkelaar et al., 2019; Chen et al., 2020b; Mcduffie et al., 2021; Weagle et al., 2018), including the concentrations of secondary inorganic aerosols, black carbon, organic aerosols, and dust. These maps were created using a combination of AOD data and in-situ PM chemical composition measurements to constrain and downscale coarse CTM outputs, via LUR techniques, to spatial scales commensurate with population density distributions. The resulting maps offered the possibility to assess the contributions of different anthropogenic emission sectors to regional and global health burden (Mcduffie et al., 2021; Chen et al., 2021b; Shi et al., 2023).

These recent developments are a fundamental first step for comprehending the differential health effects of PM components. However, models are still directly reliant on AOD and in-situ measurements and as such they cannot forecast future PM concentrations and composition and are limited in identifying the sources of the organic fraction. To address this limitation, the atmospheric science community should develop hybrid models that incorporate land-use data with CTM outputs, enabling the retention of CTMs' source-specificity and forecasting capabilities, while simultaneously benefiting from the fine-resolution information provided by land-use data. In these models, in-situ measurements should be utilized for model training, rather than as model inputs. CTM-based models have the added benefit of being able to quantify the sources of different constituents, which is especially valuable for the organic fraction. Overall, the development of hybrid models that leverage the complementary strengths of CTMs and land-use information will be key in providing exposure data suitable to determining the adverse health effects of individual PM components.

4. Field observations required to understand PM differential toxicity

In this section, we present the type of field observations required in our opinion to best capture the spatial and temporal variation of PM components, enabling validation of exposure models.

4.1 Established monitoring networks of detailed PM chemical composition

Monitoring networks play a vital role in providing essential data for understanding the spatial distribution and long-term trends of air pollution, identifying emission sources, constraining human exposure models, and evaluating the effectiveness of emission reduction measures. International

monitoring programs such as SPARTAN[2], EMEP[3], IMPROVE[4], ACTRIS[5] and ASCENT[6] have been critical in establishing and maintaining the operation of these networks. Besides the continuous provision of detailed PM measurements for policymaking, these monitoring programs offer access to outstanding facilities and openly available databases for scientists from academia and the private sector, promoting cutting-edge science and international collaborations.

Another advantage of these programs is the standardization of analytical approaches and data formats, ensuring data quality and comparability and facilitating data sharing and use. Data generated from these programs may include particle number-size distributions and the concentrations of elemental and organic carbon, major ions, and metal components. Figure 3 illustrates the distribution of stations across Europe where we have gathered detailed PM chemical properties generated from different national and pan-European programs. For some PM constituents, more than 50,000 daily concentrations at more than 200 sites are available, which is rare, if not unique. This is only possible thanks to such research infrastructures. Datasets of at least this scale are required to form a complete picture of the PM chemical and physical properties and sources, with which our atmospheric modelling community can optimize exposure maps to understand the differential health effects of PM constituents.

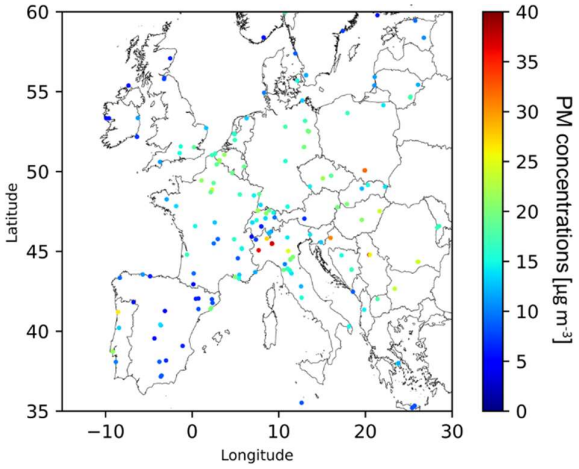


Figure 3: European map with site locations where long-term detailed chemical composition data is available. Sites are both urban and rural. Markers are colour-coded with total annual PM concentrations in 2013, to reflect differences in emissions between sites.

The composition, emission sources and formation pathways of the organic fraction remain a scientific challenge. Routine measurements (e.g., of organic carbon) are not sufficiently chemically

[2]SPARTAN: Surface Particulate Matter Network (SPARTAN) provides publicly available data on PM mass, chemical composition, and optical characteristics for connection with satellite remote sensing and for air quality management.
[3]EMEP: European Monitoring and Evaluation Programme aims to monitor and model the long-range transport of air pollutants across Europe.
[4]IMPROVE: Interagency Monitoring of Protected Visual Environments is a long-term monitoring program designed to assess the visibility and air quality in national parks and wilderness areas in the United States. The primary goal of the IMPROVE network is to measure PM mass and chemical composition, at over 170 monitoring sites across the United States.
[5]ACTRIS: Aerosol, Clouds, and Trace gases Research InfraStructure is a pan-European research infrastructure of several measurement stations across Europe that provides long-term observational data on aerosols, clouds, and trace gases.
[6]ASCENT: The Atmospheric Science and Chemistry mEasurement NeTwork is a new comprehensive, high-time-resolution, long-term measurement network in the U.S. for the characterization of aerosol chemical composition and physical properties.

resolved for the retrieval of the contributing sources. For this, two approaches are currently exploited for long-term monitoring: the aerosol chemical speciation monitor, ACSM (Ng et al., 2011; Fröhlich et al., 2013), which measures the bulk composition of the non-refractory fraction of fine PM and infrared spectroscopy, IR (Weakley et al., 2016), which measures the functional group composition of the organic fraction. We have utilized ACSM data to determine the contribution of residential emissions, vehicular emissions and secondary processes to the organic aerosol fraction across Europe (Chen et al., 2022) and to validate CTM outputs (Ciarelli et al., 2017; Jiang et al., 2019). ACSM measurements are part of ACTRIS and ASCENT, whereas the IMPROVE network has adopted IR measurements. The complex composition of the organic aerosol, especially of the oxygenated secondary fraction, means that no technique is complete. The spectra acquired with both ACSM and IR techniques retain information on the source origins and the formation pathways of the organic fraction. These two techniques are complementary and their combination, although currently only exploited in the laboratory (Yazdani et al., 2021, 2022), can be very powerful to further characterize the organic aerosol fraction in dense networks over long-terms, enabling a better understanding of the relationship between its composition and health effects.

Overall, it is essential that the scientific community continues to leverage chemically-speciated PM data from monitoring networks and generates additional datasets for validating exposure models required to understand PM differential toxicity. These networks should include both urban and rural locations, which is key for accurately characterizing PM concentrations and chemical composition in the different types of areas people reside. It is also vitally important that governments continue investing in monitoring programs to ensure the continuous acquisition of detailed air quality data.

4.2 Why detailed atmospheric chemistry matters: a comparison of severe PM pollution in Northern China and Northern India

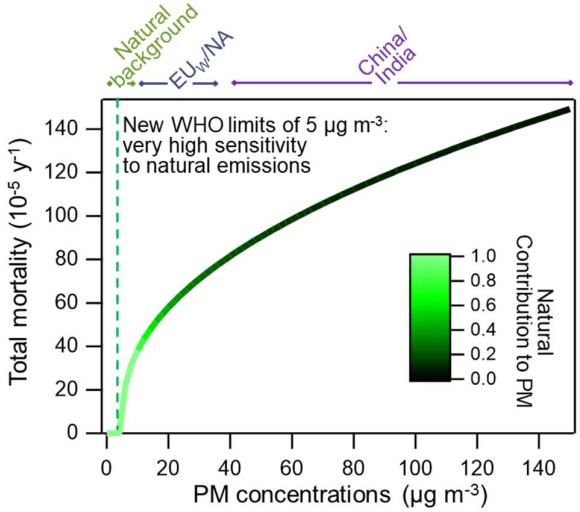


Figure 4: Dose-response relationship between PM concentrations and total attributable mortality, highlighting the sensitivity of mortality to reductions in anthropogenic emissions at low and high pollution levels and potentially to contributions from natural emissions. Reproduction adapted from (Apte et al., 2015). Vertical axes indicate per-capita mortality rates attributable to $PM_{2.5}$ for a hypothetical global population uniformly exposed to a given level of $PM_{2.5}$. The dose-response relationship is coloured by the contribution of natural emissions to PM mass. For illustration, we have chosen a natural background concentration of 5 µg m^{-3}, representing the level to which 50% of the global population would be exposed if all anthropogenic emissions were eliminated (Pai et al., 2022).

The horizontal bars at the top of the figure represent typical PM concentrations in Western Europe/north America (EU_W/NA) and China/India, as well as natural background PM concentrations.

Modest improvements in PM pollution in relatively clean regions in Western Europe and North America, where most of the current monitoring programs operate, would result in large avoided mortality, owing to the nonlinear concentration-response relationships (Apte et al., 2015). By contrast, major improvements in air quality would be required to substantially reduce mortality in more polluted regions, such as China and India (Figure 4), although such improvements are at least possible as high concentrations result from more controllable anthropogenic emissions. In China and India, 5 million deaths every year are related to air pollution (Lelieveld et al., 2015), with approximately 20% of the total deaths attributable to PM (Figure 5A). Projected demographic shifts in these regions indicate that in order to maintain current PM-attributable mortality rates, average PM levels must decrease by approximately 30% within the next 15 years to counterbalance the rise in PM-related deaths resulting from aging populations (Apte et al., 2015). Therefore, an effective program to deliver clean air to polluted regions is urgently needed to avoid several million deaths every year.

In response, China and India launched their country-level clean air plans in 2013 and 2019, respectively. Despite greatly improved national air quality levels compared to ten years ago (Figure 5B), China is now finding further air pollution reduction challenging due to the trade-off between controlling PM and ozone pollution (Li et al., 2019). The situation in India is more alarming, with the country's air quality continuing to worsen despite the implementation of its clean air program. A growing number of cities experience severe pollution (Ghildiyal, 2022), resulting in a rise of the mortality attributable to PM (Figure 5B). The mechanism of haze formation in the two regions is also very different. While pollution in China happens on regional scales, local pollution in India plays a prevailing role. The comparison between severe PM pollution in Northern China and Northern India serves as a perfect example for why a detailed understanding of the complex atmospheric chemistry involved is required to comprehend PM differential toxicity in those regions.

In China, secondary aerosol production was identified as the main cause behind winter haze events in a study conducted by Huang et al. (2014), the first of its kind to make this discovery a decade ago (Huang et al., 2014). Later studies have confirmed that in Chinese megacities, particle formation, often observed at the onset of haze, is driven by the photochemical production of secondary organic and inorganic species, which happens on a regional scale during the day (Yao et al., 2018; Kulmala et al., 2021). Multiphase chemistry plays another essential role in aerosol formation during haze in China. The high concentrations of anthropogenic sulfate and nitrate, coupled with high relative humidity, provide a reactive medium for heterogeneous aerosol production (Tong et al., 2021). Under haze conditions, gaseous NO_2 may also act as an oxidant in the concentrated aqueous aerosol media producing additional secondary PM (Su et al., 2020; Wang et al., 2020). Compared with gas phase photochemical reactions, which are self-buffered against heavy pollution, multiphase chemistry exhibits a positive feedback, which means that higher PM levels accelerate multiphase production, further increasing PM concentrations (Le et al., 2020). Furthermore, because of the nonlinear chemistry of ozone production and titration in winter, the recent reductions in nitrogen oxides result in ozone enhancement in urban areas (Li et al., 2019), further increasing the atmospheric oxidation capacity and facilitating secondary aerosol formation (Le et al., 2020). Substantial oxidation in China's atmosphere is at play even during the night. New findings reveal that between 2014 and 2019, the decrease in pollution has led to an increase in the production rates of nitrate radicals across China, suggesting the growing role of nighttime chemistry to China's air pollution (Wang et al., 2023a). Further mitigating air pollution and its health effects in China will require a detailed mechanistic understanding of the

complex multiphase and oxidation chemistry involved, as well as the identification of the major sources of secondary aerosol precursors.

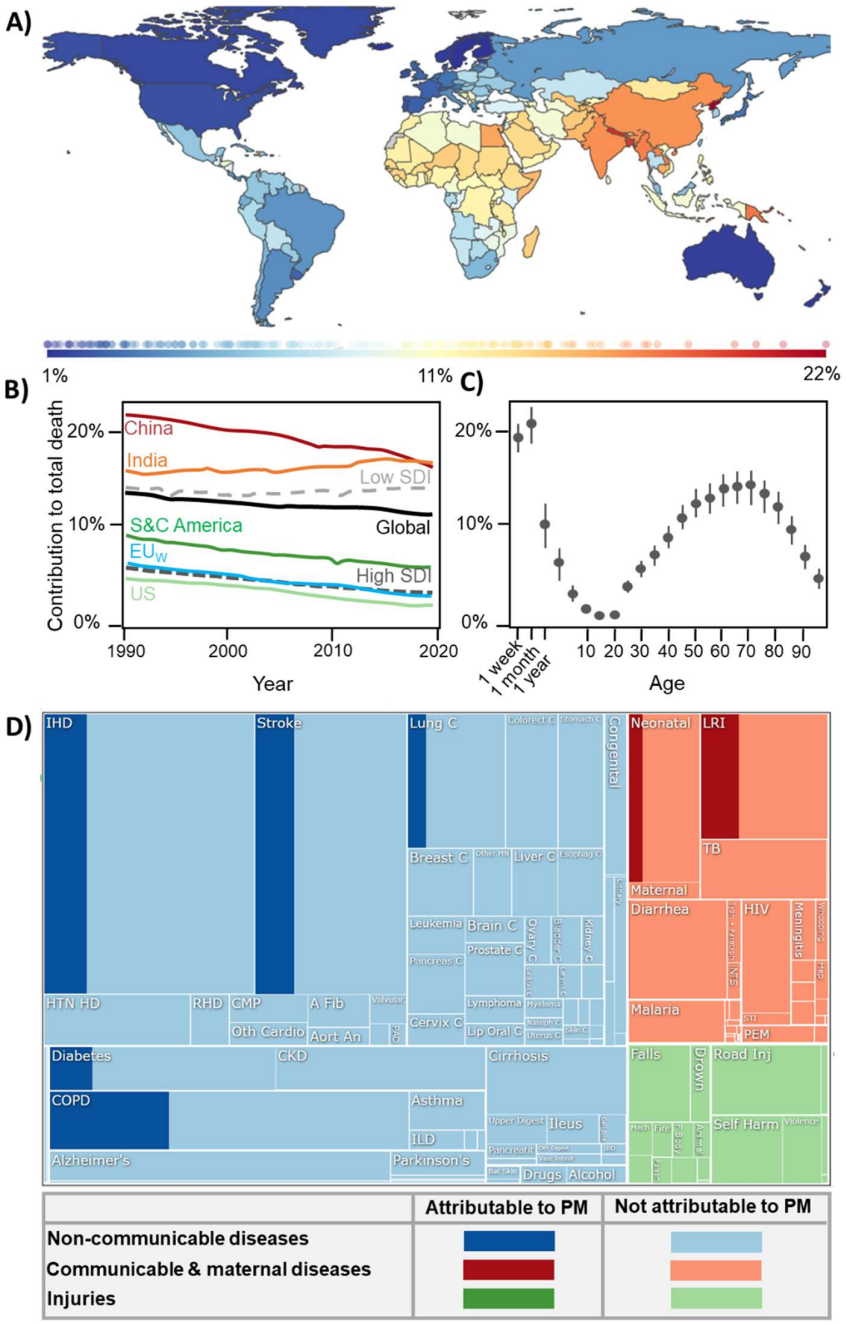


Figure 5: percentage of mortality attributed to particulate pollution. Data are from the Global Burden of Disease Study 2019 Results (Seattle, United States: Institute for Health Metrics and Evaluation, 2020 - available from https://vizhub.healthdata.org/gbd-results/). A) Percentage of PM-related mortality for every country. B) Evolution of the percentage of PM-related mortality from 1990 to 2019 for locations discussed in the text, including China, India, Western Europe (EU_W), US, South and Central America, low SDI (Socio-demographic Index) and high SDI. C) Percentage of PM-related mortality globally as a function of age. D) Percentage of deaths attributable to PM pollution related to non-communicable diseases, communicable & maternal diseases and injuries. The main causes of attributable death to which PM exposure contribute include ischemic heart diseases (IHD), stroke, diabetes, chronic obstructive pulmonary diseases (COPD), neonatal infections, and lower respiratory infections (LRI).

In Delhi-India, however, the rapid growth of particles into sizes relevant for haze formation occurs during nights without any photochemistry. We have recently shown that the growth of sub-100 nm particles is predominantly driven by primary supersaturated organic vapors from local biomass combustion emissions, whose condensation is promoted by the rapid decrease in air temperature and the increase in emissions during nighttime (Mishra et al., 2023). The formation of ammonium chloride enhances aerosol water uptake through co-condensation at high nighttime relative humidity, which sustains particle growth at higher sizes (Mishra et al., 2023) and leads to fog formation and a 50% reduction in visibility (Gunthe et al., 2021). This process, apparently unique to India's capital, does not involve photochemistry but is instead driven by high emissions of hydrochloric acid, possibly from local industries (Rai et al., 2020). During daylight hours, with the dispersion of NO_X emissions and the increase in the atmospheric oxidation capacity, local combustion of fossil fuels and biomass become an important source for SOA production (Kumar et al., 2022). Toxic heavy metal pollution levels in Delhi are another cause for alarm, with concentrations several hundred times higher than those found in Europe, also due to local industries (Rai et al., 2021). Solving air pollution in India will require international collaboration with local researchers to better understand the local sources of different pollutants, e.g. through mobile measurements (Section 4.3), as well as the effects of local meteorological conditions on air quality. Given the significance of local pollution sources, it will also necessitate the involvement of social scientists and local communities to induce social changes and raise public awareness.

The atmospheric science community has already made significant strides in understanding the sources of air pollution in China and India, but knowledge gaps still exist. It is imperative to further understand the non-linear effects of emissions on the atmospheric oxidation capacity, particularly in light of India's potential to face the same problems as China in the near future when primary pollution reduction will lead to an increase in the photochemical production of ozone and secondary aerosols. It is also crucial to identify on a molecular level the specific ingredients contributing to aerosol formation and growth and relate these ingredients to the emission sources of their precursors. We also need to gain a mechanistic understanding of the interplay between the soluble inorganic fraction and water and their effects on the enhanced partitioning and heterogeneous chemistry of organic and inorganic vapors (e.g. N_2O_5, HCl, HNO_3, and oxidized organics). Without this knowledge, we cannot accurately predict the fate of these vapors with future reductions in the anthropogenic emissions of inorganic precursors (SO_2 and NO_X). Detailed knowledge of atmospheric chemistry is key for understanding PM chemical composition and differential toxicity in these polluted regions.

Finally, it is essential to establish national monitoring networks in both countries that probe the spatial distribution and long-term trends of air pollution and allow us to evaluate the effectiveness of emission reduction measures. The data resulting from these monitoring programs serve as a cornerstone for understanding the health effects of the PM components specific to China and India, enabling us to devise regionally specific solutions aimed at effectively limiting air pollution in these regions. More generally, the inequity of air pollution is flagrant, with locations having a low socio-demographic index (SDI) suffering three times the burden of PM-related mortality compared to locations with high SDI (Figure 5B). This disparity underscores the urgent need for comprehensive monitoring programs in low SDI countries.

4.3 Urban mapping of PM chemical composition

Monitoring networks have limited spatial coverage, which can make it difficult to capture localized pollution hotspots, especially from primary combustion emissions (Eeftens et al., 2012b; Elser

et al., 2016; Elser et al., 2018; Jedynska et al., 2015; Jedynska et al., 2014). Therefore, several approaches have been proposed to enhance the spatial coverage of urban pollution measurements (Figure 2). Both ground-based sensor networks, e.g. for CO_2, black carbon, NO_2, or total PM (Popoola et al., 2018; Caubel et al., 2019; Oney et al., 2015), and satellite retrievals (Di et al., 2016; Griffin et al., 2019) can map the concentrations of individual pollutants at sub-km-scale resolutions, however, these approaches lack the chemical resolution needed for the measurements of PM composition. Aircraft measurements are suited for studying pollution plumes at regional scales (Fry et al., 2018; Decker et al., 2019), but cannot access fine scale variations at the ground level. Ground-based mobile laboratories can house online instrumentations that provide high chemical resolution, while operating with sufficiently high time resolution (i.e., few minutes) for measurements at street levels (Shairsingh et al., 2018; Gu et al., 2018). This makes them ideally suited for spatial mapping of PM composition in urban environments (Hankey and Marshall, 2015; Alexeeff et al., 2018; Apte et al., 2017; Gu et al., 2018).

A large number of studies have measured black carbon, NO_2, total PM mass and number concentrations aboard of mobile platforms (Alexeeff et al., 2018; Apte et al., 2017; Hankey and Marshall, 2015; Shairsingh et al., 2018; Simon et al., 2017; Miller et al., 2020). The Aerodyne aerosol mass spectrometer (AMS) has also been used for the mobile measurements of non-refractory PM components, including secondary inorganic species and organic aerosol (Elser et al., 2016; Elser et al., 2018; Gu et al., 2018; Mohr et al., 2011; Shah et al., 2018). The application of factorization techniques to the measured organic mass spectra has enabled its apportionment to primary traffic, cooking and biomass burning emissions as well as the quantification of a total secondary fraction (Gu et al., 2018; Elser et al., 2016; Elser et al., 2018). From measurements in the EU and the US, it was found that the secondary organic and inorganic fractions are homogeneously distributed across cities, while primary emissions are enhanced by several $\mu g\ m^{-3}$ compared to background levels (Elser et al., 2016; Elser et al., 2018) in correlation with land-use variables (Gu et al., 2018).

Until recently, there has been no robust technology for highly time resolved measurements of airborne particulate metals. Therefore, studies had previously relied on integrated offline samples collected over days-to-weeks at few sampling stations in order to assess the spatial distribution of yearly particulate metals across cities (Li et al., 2016a; Van Ryswyk et al., 2017; Zhang et al., 2015) and countries (Chen et al., 2020b). With such measurements, intra-urban variability in metal concentrations can still be discerned. Recently, the Xact 625 ambient metals monitor, an online XRF spectrometer, has been developed and successfully deployed in the field for the real time measurements of particulate elements (~25) with time resolutions down to 30 minutes (Furger et al., 2017). Due to its high temporal resolution, sensitivity and robustness in the field, the Xact is capable of delivering several month long datasets of 1000s of data points – 10-100 times more than offline techniques (Manousakas et al., 2022), which allow the retrieval of daily concentration patterns. However, further developments are needed to achieve particulate elemental analysis on timescales of minutes suitable for mobile measurements. The availability of such measurements will be transformative, enabling access to PM elemental composition at unparalleled resolutions, and the development of more robust exposure models for metal components.

Street-level PM composition data can enhance, challenge, or confirm various air quality datasets used to retrieve PM differential toxicity, such as CTM outputs, land-use regression predictions, and remotely sensed observations. This refinement can also aid addressing the effect of human mobility in epidemiological studies (Zeger et al., 2000).

5. Gaps in understanding emissions

Human activities have profoundly altered the earth's environment, impacting emissions, atmospheric composition, global temperatures, and land cover. In Figure 6, we categorize the complex anthropogenic effects on PM composition into four broad classes:

- **Direct emissions:** encompassing anthropogenic PM and PM precursors directly released into the atmosphere.
- **Land-use changes:** including changes in urban infrastructure, green initiatives, deforestation/forest management, and agricultural practices, affecting emissions and their accumulation patterns.
- **Direct effects of anthropogenic emissions on the chemistry of natural PM:** whereby pollutants from human activities react with biogenic emissions leading to PM formation.
- **Indirect perturbation of natural PM:** through anthropogenic emissions that impact natural ecosystems, such as global warming, increased CO_2 concentrations, shifts in vegetation patterns, or desertification.

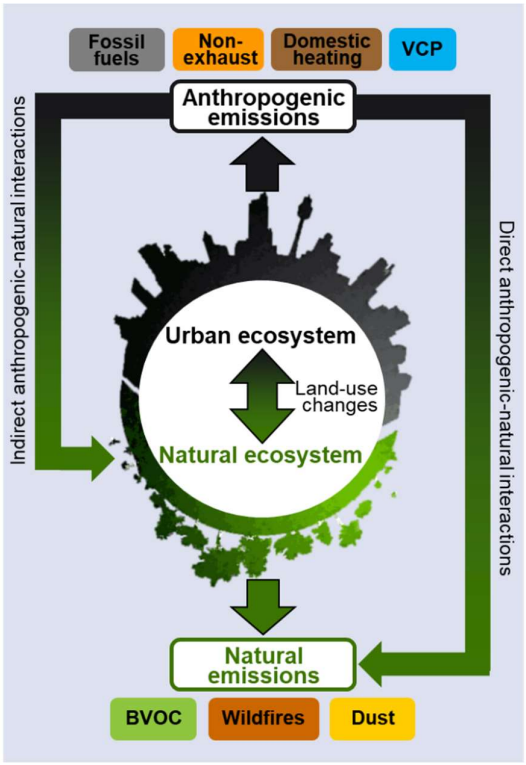


Figure 6: Anthropogenic effects on PM through (1) direct emissions, (2) land-use changes, (3) direct and (4) indirect perturbation of natural PM. (3) comprises the direct effects of anthropogenic emissions on the chemistry of natural aerosols, while (4) describes the influence of anthropogenic emissions on natural ecosystems, e.g. through global warming, or increase in CO_2 concentrations. Natural emissions from terrestrial systems include biogenic volatile organic compounds (BVOCs), wildfire emissions and dust. Anthropogenic emissions include NO_X and SO_2 from fossil fuel combustion, non-exhaust emissions, solid fuel combustion for domestic heating, and volatile chemical products (VCPs).

This section addresses existing gaps in understanding anthropogenic emissions, their atmospheric transformation, and their direct and indirect influence on natural PM. It is crucial for the atmospheric science community to approach these gaps from a mechanistic standpoint and incorporate them into models to accurately quantify the anthropogenic impacts on PM composition and thereby health effects. In section 5.1, we discuss anthropogenic PM sources that hold relevance for public health,

while in section 5.2, we examine the future trajectory of the natural PM background and its interactions with anthropogenic activities.

5.1 Legacy and emerging anthropogenic PM emissions

Anthropogenic emissions remain a predominant source of primary and secondary PM, posing a critical scientific and policy challenge in identifying the most harmful components to human health. Existing reviews have compiled epidemiological and toxicological evidence linking specific emissions to health endpoints (Wyzga and Rohr, 2015; Adams et al., 2015; Rohr and Wyzga, 2012; Yang et al., 2019; Morton Lippmann Lung, 2023). While ample literature covers short-term effects, especially through measurements at few stations, longitudinal epidemiological studies investigating the effect of PM chemical composition on chronic health outcomes are relatively scarce. Despite inconsistencies across studies, elemental carbon, organic aerosols, sulfate and metals have been consistently associated with increasing risks of cardiovascular and respiratory mortality and hospitalization (Chen et al., 2018a; Yang et al., 2019; Masselot et al., 2022; Wang et al., 2022; Wyzga and Rohr, 2015; Adams et al., 2015; Rohr and Wyzga, 2012; Badaloni et al., 2017; Wang et al., 2017).

We believe that the principal challenge in establishing robust epidemiological associations with specific PM components lies in their correlation with other pollutants, such as other PM components, O_3 and NO_X. Therefore, we call for improved high-resolution large scale chemically detailed exposure models that will offer the necessary variability for overcoming limitations related to correlations. Moreover, we advocate for the continual development of epidemiological multi-component methods that estimates the joint health impacts of PM components, instead of isolating the effect of individual ones. In this section, we will focus on major anthropogenic emissions, including fossil fuel emissions, non-exhaust on-road emissions, volatile chemical products (VCPs), and residential biomass burning (Table 2).

Fossil fuel combustion is an important source of sulfate, nitrate, and elemental carbon. Numerous accounts reported the higher differential toxicity of primary elemental carbon emissions, especially leading to cardiovascular morbidity and mortality (Chen et al., 2020a). Consequently, in 2021, the WHO has listed the elemental (or black) carbon (EC or BC) as one of the pollutants of emerging concern, calling for more evidence on their chronic health effects that can be used for future guidelines and regulations. For this, it is vital to provide high resolution, national and continental BC maps suited for large scale epidemiological studies (Section 3), ideally distinguishing between biomass and fossil fuel burning BC emissions (Table 2).

Sulfate and nitrate are not toxic in isolation, and their high fraction in PM and extended spatial variation complicates the determination of their health effects. Yet, the toxicity of these secondary components is perhaps indirect, through a complex multiphase interplay with other components. Sulfate, from energy production emissions of SO_2, provides an acidic medium for organic reactions, and may increase the solubility and hence the bioavailability of metal particles, potentially increasing their toxicity. Mobile emissions of NO_X have profound effects on atmospheric oxidation (Section 5.2), but also lead to enhanced partitioning (Lv et al., 2023) and subsequent multiphase reactions of soluble organic molecules, through nitrate formation. Traditionally, nitrate was considered the chemical end point of the reactive nitrogen life cycle in the atmosphere prior to wet or dry deposition. However, there has been growing evidence for particulate nitrate photochemical renoxification in the presence of light and organic molecules (Jiang et al., 2023; Bao et al., 2020). While this process is mainly examined for its potential to produce oxidant precursors (NO_X and HONO), how it alters the composition of the organic fraction is currently not understood. The mechanistic understanding of these multiphase

processes involving the interactions of secondary inorganic particles with organic and metal components is indispensable for constraining their impact on PM chemical composition and differential toxicity (Table 2). There is a need for fundamental mechanistic investigations of these processes in the laboratory and the field, especially in polluted areas, like China, where multiphase chemistry plays a key role for haze formation (Section 4.2).

As traffic exhaust emissions of NO_X, PM and hydrocarbons are increasingly regulated, car engines underwent a technological revolution, improving combustion efficiency and after-treatment technologies. In contrast, non-exhaust emissions, such as brake and tire wear, have increased with the growing number of vehicles and currently exceed exhaust emissions (Timmers and Achten, 2016). These emissions include toxic metals such as copper, which enhance PM oxidative potential (Daellenbach et al., 2020). Even with the electrification of the fleet, non-exhaust emissions will remain an issue, potentially worsened by the heavier weight of electric cars (Timmers and Achten, 2016). While public transportation, including trams and trains, may also be an important source of metal particles, their contribution is not yet well quantified. Multi-site long-term measurements (Section 4.1) and mobile measurements to map trace element concentrations (Section 4.3) may offer new opportunities to represent the distribution of on-road non-exhaust emissions in exposure models (Table 2).

Table 2: Future changes in anthropogenic emissions, key observations needed for coupling with health data, high priority model developments for understanding the health effects of anthropogenic emissions and their future evolution.

Source	Future changes	Key observations	Model developments
Fossil fuel combustion	Decrease by several % per year of SO_2, NO_X and BC in the West and China.	Long-term, multi-site measurements of BC, ammonium sulfate and nitrate for model improvements. Mobile measurements of BC in urban and rural locations. Apportionment of BC between fossil and non-fossil emissions. Fundamental studies and field observations of the multiphase interactions between ammonium sulfate and nitrate with the organic and metal components.	Fine resolution modelling of BC concentrations, ideally from different combustion sectors. Implementing the effects of nitrate and sulfate on the organic and metal components and estimating future changes with decreasing fossil fuel emissions.
Non-exhaust emissions	Increase of the total burden by several % per year with the increase and electrification of the vehicular.	Long-term, multi-site measurements of elements, with a focus on copper. Mobile measurements of trace elements in urban and rural locations.	Fine resolution modelling of PM elemental composition, with a focus on copper. Utilization of long-term trends for separating between the effects of exhaust and non-exhaust emissions. .
Volatile chemical products (VCPs)	Increase of the total burden by several % per year with the increase and westernization of the global population.	Identification and multi-site measurements of VCPs and VCP SOA markers. Determination of SOA formation potential of individual and real-world mixtures of VCPs.	Modelling of SOA and ozone formation from VCPs on continental levels for exposure assessment.
Biomass burning for residential heating	Area dependent.	Long-term, multi-site measurements of biomass burning primary and aged emissions. Mobile measurements of primary and aged biomass burning emissions in urban and rural areas. Fundamental studies of biomass smoke aging.	Fine resolution modelling of biomass burning emissions. Implementing biomass burning aging mechanisms in models. .

With the drastic reduction of on-road transportation emissions, VCPs, which are partly from indoor emissions, have emerged as one of the largest sources of outdoor urban organic emissions in US and European cities, modulating urban chemistry (Coggon et al., 2021; Gkatzelis et al., 2021; Mcdonald et al., 2018). These ubiquitous emissions encompass pesticides, coatings, printing inks, adhesives, cleaning agents, and personal care products. Human exposure to fossil carbonaceous aerosols may be transitioning from transportation-related sources to VCPs. These emissions have comparable, if not greater, SOA potentials compared to vehicular emissions, which may influence human health. Variations in SOA potentials and chemistry among VCPs, as revealed by laboratory experiments, highlight the need for further characterization of these unconventional emissions (Shah et al., 2020). Furthermore, it is now possible to include these emissions into exposure models (Pennington et al., 2021). Existing regulations on VCPs emphasize reducing ozone and air toxics, but currently exempt numerous chemicals that contribute to SOA formation. Efforts to refocus mitigation strategies require atmospheric scientists to provide data quantifying the contribution of VCP emissions to SOA, and the global burden of disease (Table 2).

Achieving net-zero emissions for climate goals does not necessarily guarantee clean emissions for air quality. Biomass combustion, adopted as a carbon neutral energy source for residential heating, is a potent anthropogenic source of pollution during winter. The emitted organic vapours rapidly react in the atmosphere with OH and NO_3 radicals, resulting in substantial SOA production (Kodros et al., 2020; Stefenelli et al., 2019). The SOA formed contains high levels of oxygenated and nitro-aromatic compounds, which likely cause the high oxidative potential of this fraction (Daellenbach et al., 2020). Recent laboratory investigations (Liu-Kang et al., 2022; Wang et al., 2023b) and airborne field measurements (Morgan et al., 2020; Zhou et al., 2017) suggest that primary biomass emissions, which absorb near UV light, can undergo photoreactions in the particle phase, resulting in a doubling of the emissions oxidation state in few hours. The dominant transformation processes of biomass burning emissions and their impact on aerosol toxicity remain unclear. Overall, biomass smoke has not shown a reduction trend in many regions worldwide, underscoring the importance of comprehending the fate of these emissions in the atmosphere and their implications for human health (Table 2).

5.2 Anthropogenic effects on natural PM

With the increasing regulations on anthropogenic emissions, the contribution of natural emissions, including biogenic volatile organic compounds (BVOCs), wildfires and desert dust, will gain prominence (Figure 4). While these emissions stem from natural ecosystems, they are also significantly perturbed by anthropogenic activities, as illustrated in Figure 6. The traditional picture that distinguishes biogenic and anthropogenic sources obscures human impacts on ostensibly natural systems. Anthropogenic effects on natural PM can be either direct, through the alteration of atmospheric reactivity, or indirect, through feedback mechanisms triggered by changes to the biosphere. We need to understand these effects quantitatively to devise best practices to mitigate their impacts. For example, WHO good practice statement on particles originating from sand and dust storms (SDS in the WHO 2021 air quality guideline document) includes measures that can be implemented to mitigate exposure. In this section, we discuss the human influence on natural PM concentrations, chemical composition, and future trends (Table 3).

Biogenic SOA (BSOA) is the most important source of OA in the atmosphere (Jiang et al., 2019), with anthropogenic NO_X playing a vital role in moderating its formation and composition. NO_X effects on BSOA are multifaceted and involve (1) altering the fate of biogenic RO_2 radicals, (2) increasing the atmospheric oxidant concentrations and (3) providing an aqueous medium for additional

multiphase reactions (Xu et al., 2015; Pye et al., 2019; Carlton et al., 2018). As NOx emissions decrease, RO_2 autoxidation becomes increasingly important, potentially enhancing BSOA formation, while oxidant availability driving RO_2 formation rates simultaneously declines, possibly slowing regional BSOA formation. Recent modelling analyses (Carlton et al., 2018), along with in-situ (Xu et al., 2015) and airborne measurements (Pye et al., 2019; Shrivastava et al., 2019) consistently suggest that anthropogenic NO_X leads to a net enhancement in BSOA concentrations by 20-50% depending on the location and season. Like NO_X, SO_2 emissions from electricity generation, the main source of particulate sulfate, modulate the aqueous formation of isoprene SOA. Models (Carlton et al., 2018) and measurements (Xu et al., 2015) over the US demonstrate that between 40–70% of the BSOA can be controllable by reducing anthropogenic NO_X and SO_2. Similar analysis is still lacking at other locations worldwide.

Table 3: Future changes (2050-2100) in natural emissions, key observations needed for coupling with health data, high priority model developments for understanding the health effects of emissions and their future evolution.

Source	Future changes	Key observations	Model developments
Biogenic SOA	Increase in global BSOA burden by 30-150%.	Long-term, multi-site measurements of BSOA precursors, oxidant precursors, chemistry, and burden. Global analysis of response of BSOA chemistry and burden to anthropogenic emissions and climate change. Fundamental studies of anthropogenic-biogenic interactions and their effects on BSOA chemistry and burden.	Improving the understanding of the response of BVOC emitting species to climate change (temperatures, soil nutrients, CO_2, nitrogen deposition, droughts, vegetation shifts). Implementing the effects of anthropogenic-biogenic interactions on BSOA chemistry and burden
Wildfires	Increase in wildfires frequency by ~100% and emission burden by ~30%.	Long-term global records of fire occurrence and associated PM emissions. Global analysis of response of wildfire emission occurrence and budget as function of climate change, and fire drivers (temperature, droughts, lightning). Determination of wildfire emission rates for different ecosystems. Fundamental studies of wildfire emission and their atmospheric transformation processes.	Coupling fire and vegetation models. Improving the understanding of the impact of land and fire management on fire emissions.
Dust	Uncertain	Long-term global records of dust emission burden, size and chemical composition. Quantification of the contribution of soil vs. urban dust in major cities. Field and laboratory observations of dust aging and its impact on the bioavailability of key elements.	Improving dust emission schemes. Implementation of dust updated aging schemes.

BSOA concentration exhibits a strong temperature dependence, driven by the exponential increase in BVOC emissions and their oxidation rates. Our analysis of multi-field observational datasets from European and North American locations reveals that BSOA contributes [0.9–2.5] μg m^{-3} at 15°C, compared to [2.1–6.3] μg m^{-3} at 25°C (Xu et al., 2015; Daellenbach et al., 2017). Climate models project a global increase in BSOA mass by approximately 30–150% with a temperature rise of 2°C and a few hundred ppb increase in atmospheric CO_2 concentrations (Carslaw et al., 2010). When changes in vegetation are accounted for, predictions of BVOC emissions become extremely uncertain, with projected increases ranging from 10s to 100s of percent (Table 3). These uncertainties arise from the unpredictable response of vegetation to future climates, including longer growing seasons, increased leaf area index with the fertilization effect of CO_2, changes in water stress and expansion of the boreal

and temperate forests. With the rise in BVOC emissions and the denitrification of the atmosphere, it is expected that the oxidation capacity of the atmosphere may decrease leading to slower production of BSOA and a complete change in its composition. Understanding the non-linear interactions among anthropogenic emissions of oxidant precursors, greenhouse gases, atmospheric oxidation conditions, and the biosphere is crucial for understanding BSOA concentration, chemical composition, health effect and future concentration trends in different regions.

While reducing NO_X and SO_2 can control a significant portion of BSOA, the rise in BVOC emissions with climate change, albeit highly uncertain, may offset this reduction. Currently, there is very limited understanding of the impact of BSOA on human health, with only one study suggesting a 3.5 times higher cardiorespiratory mortality associated with anthropogenically-influenced BSOA compared to $PM_{2.5}$ mass (Pye et al., 2021). Atmospheric scientists should capitalize on emerging multi-year, multi-location observations of detailed PM chemistry to enhance model predictions of BSOA (Table 3).

Wildfires have become increasingly frequent in many regions worldwide, making them the second largest contributor to atmospheric organic carbon on a global scale. This source can be directly affected by anthropogenic activities, through deforestation, forest management and fire suppression or indirectly by climate change. Climate models predict that global warming will amplify wildfire emissions by ~30%, owing to longer fire seasons, higher temperatures, increased droughts, and increased convection-induced lightning as an ignition source (Carslaw et al., 2010). Studies have shown short-term health effects of PM from wildfire emissions, including pulmonary complications (Stawovy and Balakrishnan, 2022), respiratory mortality and cardiovascular mortality (Chen et al., 2021a), while understanding the long-term health effects of these emissions remains an ongoing area of research (Grant and Runkle, 2022; Gao et al., 2023). Recent studies investigating Amazonian (Yu et al., 2022) and Canadian Boreal (Korsiak et al., 2022) wildfire emissions have highlighted an elevated risk of various cancers, surpassing the effects of non-wildfire PM emissions for equivalent exposure doses. A crucial first step in improving prediction of wildfire emissions and their health effects is the analysis of global fire occurrence records and associated PM emissions, establishing robust relationships between emissions, ecosystems, climate change, fire management and fire drivers (Table 3).

Another naturally occurring PM component, dust, is the most important source of elements in the atmosphere. Wind speed, soil moisture and vegetation cover are the main drivers of dust emission fluxes, size distribution and mineralogical composition (Carslaw et al., 2010). During transport, dust particles react with acids increasing the bioavailability of key elements, including iron. It has been shown that anthropogenic sulfate from fossil fuel combustion modulates soil dust iron solubility and toxicity (Wong et al., 2020). The reactive uptake of gases onto dust particles heavily depends on dust mineralogical composition, with particles rich in carbonates exhibiting strong atmospheric reactivity. Like wildfires, both direct human activities and climate change can influence dust emissions, making future predictions uncertain (Carslaw et al., 2010). Dust outbreaks have frequently been associated with mortality and hospital admissions (Stafoggia et al., 2016; Crooks et al., 2016), albeit with moderate effects and associated high uncertainties (Zhang et al., 2016). This uncertainty may result from the large variability in dust particles morphology, size, solubility, and chemical composition, depending on their origin and transport time in the atmosphere. A particular challenge is the provision of long-term, large-scale datasets, which is crucial because of the strong spatial and temporal variability of dust particles in the atmosphere. Therefore, additional observational data on dust phenomenology is required for model

evaluation, with which we can comprehend the toxicity of dust and the contributing factors. Finally, fundamental research on dust transformation processes is also warranted (Table 3).

While anthropogenic emissions are destined to decrease, natural emissions will most likely increase. Part of this increase can be controllable through reducing anthropogenic emissions and managing land-use. The atmospheric science community is now ready to provide the field measurements, laboratory observations and model outputs needed to quantify the contribution of anthropogenic, and controllable natural emissions globally and predict their evolution with global changes. This data will form the foundation for understanding the toxicity of anthropogenic PM sources and determining the natural PM background in different regions.

6. Supporting epidemiology by enhancing chemically-resolved PM exposure estimation

As aerosol scientists, we must shift our focus from seeking a single, elusive chemical or toxicological metric describing PM health effects to providing chemically resolved atmospheric observations and modeling outputs, with which epidemiological evidence linking PM components to diseases can be established. Epidemiology drives regulatory changes, and working with epidemiologists, atmospheric scientists can now assume a central role in determining PM composition and spatial distribution to unlock a better understanding of the complex epidemiological relationships between PM composition and health outcomes. This section highlights some notable advancements in modern environmental epidemiology that may not yet be fully appreciated within the atmospheric chemistry community.

6.1 Collaboration between atmospheric scientists and epidemiologists

Recently, eight hallmarks of environmental insults have been proposed (Peters et al., 2021). They encompass oxidative stress and inflammation, genomic mutations, epigenetic alterations, mitochondrial dysfunction, endocrine disruption, altered intercellular communication, changes in microbiome communities, and impaired nervous system function. These hallmarks jointly underpin the health effects resulting from lifelong environmental exposures, even to relatively modest concentrations of contaminants.

PM, as one of the most important environmental insults, can infiltrate our body through various barriers, e.g., our lungs or digestive system, affecting individuals through a complex web of biological pathways. Figure 5C illustrates the contribution of PM pollution to total mortality across ages, highlighting its staggering impact on infants and the elderly. Short-term exposure has been linked to sudden infant death as well as increased mortality and morbidity rates due cardiorespiratory and renal complications, particularly affecting children and individuals with chronic conditions (Heft-Neal et al., 2018; Zhang et al., 2023; Liu et al., 2023; Guo et al., 2023). Meanwhile, long-term exposures have been linked to numerous non-communicable diseases that manifest at a later stage of life (Figure 5C), including cardiovascular diseases (Requia et al., 2017; Lelieveld et al., 2019), respiratory symptoms, different types of cancers (Turner et al., 2020), diabetes (Yang et al., 2018), and neurodegenerative diseases (Maher et al., 2016; Shi et al., 2020).

Unlike infectious diseases, non-communicable diseases have multiple causes and involve various factors, which individually may neither be necessary nor sufficient to cause the disease. Early-life exposures may leave enduring marks in the body, leading to manifestations that emerge many

decades later. Given the multifactorial nature of the problem, epidemiology is irreplaceable when it comes to investigating PM-related non-communicable diseases and working with citizen cohorts is essential in circumventing the challenges of randomization and cofounding effects. Citizen involvement is simply inevitable in understanding their own health.

To bolster the causal interpretation of epidemiological associations derived from observational studies, it is crucial to elucidate how the hallmarks of environmental insults connect exposure and disease development. This requires a robust methodological approach, combining insights from aerosol science into PM chemical composition, sources and spatial distribution, with modern environmental epidemiology techniques, including molecular phenotyping, multiomics, epigenetics, imaging, and personalized and digital medicine.

6.2 Working with citizen cohorts to establish causal links

The establishment and maintenance of national biobanks and citizen cohorts is key for investigating the causal links between exposure to PM components and diseases. These cohorts are the gold standard for understanding long-term health effects of environmental factors (Probst-Hensch et al., 2022). They provide evidence where randomized trials are unethical or unfeasible (Peters et al., 2022). Cohorts are critical for approaching a causal understanding of how social, environmental, behavioral, and economic factors promote or hinder health, while also enabling the evaluation of the long-term impacts of public health interventions. They allow studying health trajectories across different ages, providing a life course perspective. As such, they serve as a fundamental pillar for addressing the health effects of PM in the context of other major public health challenges of the 21st century, including population growth, aging societies, urbanization, global warming, digital transformation and increasing social inequalities.

As highlighted in (Probst-Hensch et al., 2022), Europe has a longstanding tradition of implementing and maintaining large-scale (>100k participants) and long-term (>20 years) cohorts, including the UK Biobank (Sudlow et al., 2015), Lifelines (Stolk et al., 2008), Constances (Zins et al., 2010), and the German National Cohort (Peters et al., 2022). Innovations in these cohorts include recruitment from birth to old age, implementation of novel eHealth tools, involvements of psychologists and social scientists, and citizen participation during planning and execution to address response rate challenges. Biomaterial collection within these cohorts enables sequencing and in-depth molecular characterization, differentiating between genetic and environmental factors.

A noteworthy addition to national cohorts is the global mortality dataset, maintained by the Global Burden of Disease Collaborative Network, within the Institute for Health Metrics and Evaluation (see Figure 5). Although the dataset is limited to cause-specific mortality, this network has shaped modern epidemiology and allowed the quantification of the global burden to PM mortality (Burnett et al., 2018). Another emerging dataset is from the multi-country, multi-city network, which provides daily mortality for several locations around the world, ideal for the assessment of short-term PM exposures (Masselot et al., 2022; Liu et al., 2019). Such datasets synergistically complement the causal investigations into PM health effects based on cohort data, providing a global perspective.

Our vision is of a closer collaboration such that detailed knowledge of PM composition and exposure can be integrated into citizen cohorts, including an increasing focus on polluted areas like China and India (section 4.2). Working together will advance understanding of the involvement of specific PM components or combination of components in disease development and the detection of

early changes resulting from exposure. It will generate even more compelling evidence for science-to-citizen-to-policy partnerships needed to affect regulatory frameworks.

6.3 Disease prevention through the mitigation of detrimental PM components

It is vital to prioritize quality of life and healthy aging over simply extending life expectancy, especially in high SDI regions. This necessitates a shift towards primary prevention and the implementation of drastic changes in health promotion starting at childhood and early adulthood, well before the onset of diseases. In the case of PM, it is essential to identify and mitigate the specific components responsible for different diseases, to alleviate their impacts on our wellbeing. A reduction in detrimental PM components will also result in an extension of life expectancy, especially in low SDI locations.

Dementia serves as a perfect illustration of the major challenges facing our aging society. Dementia is a severe decline in cognitive function, which considerably affects the wellbeing of older adults and their families, while imposing substantial costs on public programs. In 2010, approximately 135 million adults were living with dementia worldwide (Prince et al., 2013), resulting in estimated economic impacts of $600 billion (Wimo et al., 2013). Given the sharp rise in dementia incidence beyond the age of 75 and our increasingly ageing society, global dementia cases are forecasted to triple by the year 2050. Recent studies in the US have shown that every 5 μg m^{-3} increase in annual PM concentrations results in a 13% increased risk of first-time hospital admissions for dementia (Shi et al., 2020), with elemental carbon and sulfate particles having the strongest effects (Shi et al., 2023). While more research, including at other locations, is necessary to confirm these associations and understand the underlying biological pathways involved, these studies already shed light on possible interventions to slow the trajectory of cognitive decline and ensure the wellbeing of our aging society. This example reinforces that a collaborative effort – combining the expertise of epidemiologists in understanding disease patterns with the experience of atmospheric scientists in measuring and modelling PM composition – is crucial for mitigating the impact of PM components on the wellbeing of our society. Such collaboration will also enable identifying the PM components contributing to other diseases, potentially operating through distinct biological pathways in their development.

7. Conclusions

In the 21st century, we have witnessed a remarkable rise in life expectancy and shifts in global disease patterns, attributable to a combination of public health interventions and advancements in healthcare and healthcare accessibility. Yet, ten million deaths attributable to environmental exposures can still be preventable every year (Neira and Prüss-Ustün, 2016; Landrigan et al., 2018), highlighting the need for proactive measures. Relying solely on high-tech medical interventions for managing disease progression may exacerbate existing social inequalities within healthcare systems and yield diminishing returns. Therefore, we advocate to shift towards enhancing quality of life and promoting healthy aging through early prevention and the creation of healthy environments for all. Our vision for realizing this goal is a close collaboration between atmospheric scientists and epidemiologists, to integrate chemically detailed global air quality data with large-scale personalized medical information from citizen cohorts. The provision of global PM composition maps will require the atmospheric science community to (1) develop spatially and chemically detailed exposure models, (2) provide long-term time-series of PM chemical composition from monitoring networks, (3) map pollution hot-spots through mobile measurements, (4) understand emerging anthropogenic emissions and their chemical

transformation, especially in heavily polluted areas like China and India, and (5) understand the future evolution of natural emissions with climate and land-use changes.

As an aggressive attempt to promote healthy environments, WHO has set new guidelines to limit PM concentrations to below 5 μg m^{-3}. Achieving these limits may be challenging for many regions due to the contribution of natural emissions from wildfires, biogenic species, and desert dust. Concurrently, scientific consensus underscores the critical role of PM chemical composition in influencing associated health effects, necessitating a revaluation of how we should be mitigating PM pollution and the development of new generation of air quality metrics focusing on detrimental PM components. Focusing on the PM differential toxicity offers two key advantages. First, it allows for targeted measures aimed to limit specific health-relevant PM sources. Second, PM chemical composition is intertwined with other properties that may also drive PM's health effects, such as solubility, number size distribution and oxidative potential. Atmospheric science has reached a pivotal moment to provide detailed global air quality maps, at a sufficiently fine resolution, supporting epidemiological studies to determine the differential toxicity of PM components, crucial for integrating PM chemical composition into regulatory frameworks, informing targeted policy-making and operational decisions.

Routine, widespread availability of high-resolution atmospheric data could have transformative implications for air quality research, epidemiology, and environmental management. This valuable data can reveal localized pollution hotspots, offering new opportunities for implementing targeted pollution control measures. When combined with personal GPS data, it enables comprehensive personalized exposure analytics, potentially influencing individual behavior. This parallels the way real-time traffic data currently shape driving patterns at an individual level or how health applications motivate individuals to engage in active exercise.

By providing open access to global high-resolution pollution maps, atmospheric scientists can assume a broader societal role in raising public awareness of air pollution, and its impacts on health and environmental equity. These pollution maps empower citizens, local communities and policy makers with the necessary tools to optimize emission reduction strategies and sustainable urban planning. This can include the application of targeted measures for limiting the most important PM sources for health, or shifts in urban land-use design for better air quality. This wealth of data can be utilized to train models that predict the future evolution of air pollution sources and its health impacts with climate change, land use change, urban planning, mitigation strategies and energy policies. Long-term global air quality data are a key cornerstone for establishing targeted strategies to improve public health and anticipate its future trajectory.

In the process of reevaluating and implementing air quality guidelines, a multidisciplinary collaborative approach involving atmospheric scientists, climate scientists, toxicologists, epidemiologists, public health experts, social scientists, policy-makers, and the public is crucial. Therefore, governments must ensure sustainable funding to foster these collaborations, the returns in terms of lives and costs saved being increasingly evident. By alleviating the burden of air pollution-related diseases, we will prioritize the health and wellbeing of individuals and create sustainable and resilient communities.

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