Solid Particulate Matter in the Atmosphere

Reto Gieré[1] and Xavier Querol[2]

Atmospheric particulates—tiny particles in the air—represent an exciting new research area for mineralogists and geochemists. Emitted directly into or formed within the atmosphere, these particles are generated by both natural processes and human activity. Although derived mostly from sources that are spatially and temporally confined, the particles are ubiquitous globally due to atmospheric circulation. Depending on their physical and chemical properties, these small particles have local- to planetary-scale environmental impacts, influencing the radiative properties of the atmosphere and the cryosphere, the nucleation of both warm and ice clouds, and the nutrient contents of oceans and soils. Because airborne particles can affect human health and transportation, mainly aviation, they have become a focus of government attention and regulation.

KEYWORDS: aerosol particles, single-particle composition, climate forcing, cloud formation, ecosystem fertilization, aviation

INTRODUCTION

Solid particles and liquid droplets are ubiquitous in the atmosphere and are collectively referred to as particulates or particulate matter (PM). In contrast, the term *aerosol* refers to both the PM and the gas in which it is suspended. Because of the great variability in their chemical, physical, and mineralogical properties, and in their spatial and temporal distribution, atmospheric particulates constitute one of the major uncertainties in climate-change models. In this issue of *Elements,* which focuses on solid atmospheric particles, we will explore the atmosphere as an exciting new research area for mineralogists and geochemists, one with fundamental importance for human well-being. Research carried out in this general field is aimed at understanding key interactions between the solid Earth and its hydrosphere, biosphere, atmosphere, and the anthroposphere.

Humans have been aware of airborne particles since antiquity, as documented for example by records of the eruption in 79 CE of Mount Vesuvius. In his letter to the historian Tacitus (*Epistulae* VI, 16), Pliny the Younger described an ash cloud of "unusual size and appearance" and shaped "like an umbrella pine." Pliny further suspected that the death of his uncle during this catastrophe was caused by his breathing being "obstructed by the dense, dust-laden air." Dust also fascinated Christian Gottfried Ehrenberg (1847), the father of micropaleontology, who carried out detailed microscopic and geochemical investigations on reddish-brown dust and "blood rain." Since ancient times, and as described in Homer's *Iliad* (Book 16, 458-461) and later in the Roman literature, these unusual natural phenomena have been considered both intriguing incidents and divine omens. Ehrenberg observed that some atmospheric dusts contain abundant particles derived from living organisms. Using characteristic species of algae, phytoliths, cysts, and spores, he tried to determine the provenance of dusts deposited on Europe and the Atlantic Ocean. The apparent absence of typical African species in dust collected on Charles Darwin's *Beagle* near the Cape Verde Islands led Ehrenberg to postulate that the material did not originate in Africa. This conclusion was contested by Darwin (1846), who proposed that the dust was transported to his vessel by the *harmattan*, a hot West African trade wind. Darwin's interpretation turned out to be correct, as impressively revealed by modern satellite imagery (FIG. 1).
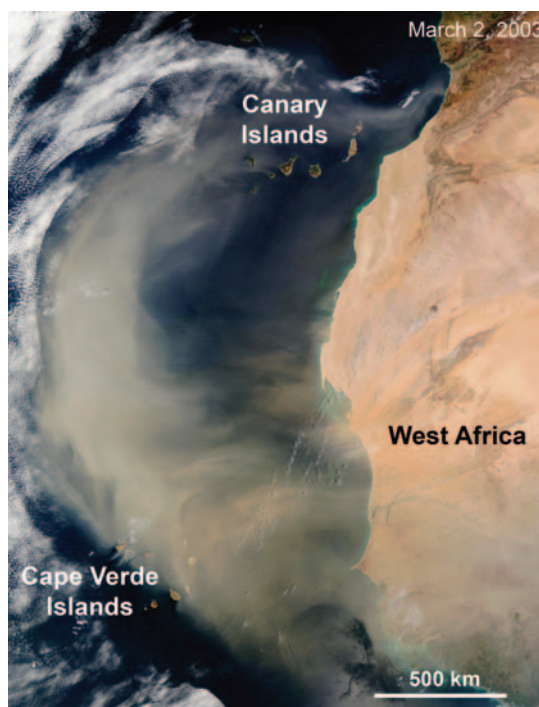
1 Institut für Geowissenschaften, Albert-Ludwigs-Universität
   79104 Freiburg, Germany
   E-mail: giere@uni-freiburg.de

2 Institute of Environmental Assessment and Water Research (IDÆA)
   C/LLuis Solé i Sabarís S/N, 08028 Barcelona, Spain
   E-mail: xavier.querol@idaea.csic.es

FIGURE 1   Saharan dust plume extending more than 1600 km over the Atlantic Ocean off West Africa.
IMAGE COURTESY OF NASA

The sources of many other types of atmospheric particles, however, have never been disputed. In medieval Europe, for example, black carbonaceous particles ("soot") have long been known to result from coal combustion. Recognizing that, for centuries, black smoke was affecting the health of his subjects, King Edward I of England declared that burning coal would be punishable by death, and at least one man was executed for violating what may have been the first air pollution law (Rottenberg 2003). Despite further attempts to curb the usage of coal in the 14th and 15th centuries, smoke pollution kept increasing, especially during the industrial revolution. In the Victorian era, English factory towns were characterized by the presence of "interminable serpents of smoke," the words used by Charles Dickens in his novel *Hard Times* (1854). During one of history's most infamous air pollution episodes, London's "Great Smog" or "Big Smoke" in December 1952 (Fig. 2), a dense mixture of smoke and fog (hence called *smog*) accumulated beneath a well-developed temperature inversion. It persisted for five days and caused approximately 4000 deaths from respiratory and related medical conditions (Stone 2002).

These historic examples document the diversity of atmospheric particles in terms of material, source, distribution, and impact. In the following sections, we provide additional details on these topics and define some key terms.

TYPES AND SOURCES

Particulates emitted directly into the atmosphere are known as *primary particles* (e.g. mineral dust), whereas those formed within the atmosphere through gas-to-particle conversion are termed *secondary particles* (e.g. ammonium nitrate). Once particles are suspended in the atmosphere, they may be altered or removed, so that atmospheric residence times may vary from a few days to several weeks. PM occurs in both the troposphere and the stratosphere, regions separated by the tropopause, which is located at an altitude of ~9 km in the polar regions and ~17 km in the tropics.

In the *troposphere*, the bottom layer of the atmosphere, various types of naturally generated PM can be distinguished. These include particles resulting from explosive

volcanic eruptions, bursting of bubbles at the sea surface, dust storms, and forest and peat fires (Figs. 3, 4). They also comprise biogenic particles, which include plant debris, animal fragments, pollen, spores, algae, fungi, bacteria, and viruses, and also secondary organic aerosol (SOA) particles formed in the atmosphere from biogenic volatile organic compounds (VOCs). The troposphere also contains anthropogenic PM resulting from industrial operations, construction and demolition processes, traffic, surface mining, metal smelting, waste incineration, and combustion of fossil fuels and biomass.

In the *stratosphere*, PM is derived from extraterrestrial material and large explosive volcanic eruptions (ash particles), and also from the oxidation of carbonyl sulfide (COS) and volcanic sulfur dioxide (SO_2), producing sulfuric acid (H_2SO_4) particles. Emitted into the troposphere mainly from the oceans, COS can reach the stratosphere, where it is probably the main contributor to the sulfate layer during periods of low volcanic activity (Crutzen 1976). In general, stratospheric PM is characterized by lower particle concentrations, fewer particle types, and longer residence times than tropospheric PM.

SIZE DISTRIBUTION

Size is one of the most important parameters used to describe PM properties and behavior. Even though many particles are not spherical, size is typically classified using their diameter (d) which, in many cases, represents the equivalent diameter of a spherical object with some of the same physical properties (Buseck and Schwartz 2004). Atmospheric PM exhibits a considerable range in diameter, from a few nanometers (nm) (representing the smallest clusters of atoms or molecules that can be classified as particles) to several tens of micrometers (µm) (the largest particles that can be suspended in air). Particles with $d <$ 2.5 µm ($PM_{2.5}$) are generally described as *fine* and those with larger diameters as *coarse* particles (Fig. 5). This distinction is essential because the two size ranges, or modes, are fundamentally different in terms of formation, modification ("ageing"), removal, atmospheric residence times, and physical and chemical properties (Seinfeld and Pandis 2006), as well as in regard to locus of deposition in the human respiratory tract (Newman 2001). It should be noted that the boundary between fine and coarse particles at 2.5 µm was defined on a regulatory rather than scientific basis. It does not coincide with the volume-distribution minimum that separates particles formed by different processes (Fig. 5), and therefore some authors prefer a value of 1–2 µm for this boundary. Throughout this issue of *Elements*, we use the value of 2.5 µm for the upper limit of the fine-particle mode.

Fine PM is further subdivided into three size fractions to account for distinct characteristics. Particles with $d <$ 10 nm (known as *nucleation-mode particles*; Fig. 5) may form through condensation of hot combustion vapors (e.g. flue gas) or homogeneous nucleation of atmospheric gases. Particles with d = 10–100 nm belong to the so-called *Aitken mode* and form mostly through coagulation of nucleation-mode particles or condensation on them. Particles belonging to these two modes are the most abundant but, due to their minute size, account for only a minor proportion of the total PM volume or mass (Fig. 5). The third and largest fine-size fraction, the *accumulation mode,* comprises particles generated directly as primary emissions, by condensation of low-volatility vapors on existing PM, and by coagulation of smaller-mode particles. Coagulation is a relatively slow process, which generally prevents fine particles from growing larger than approximately 1 µm (Whitby 1978). Particles in the accumulation mode therefore range

in diameter from 0.1 to ~1 µm (Williams and Baltensperger 2009), but the upper limit is also described as ~2 µm or ~2.5 µm (Jacobson 2002; Seinfeld and Pandis 2006). For simplicity, we have adopted and have used throughout this issue of *Elements* the value of 2.5 µm as the upper limit for the accumulation mode (consistent with the regulatory upper limit for fine particles, as noted above). Particles in the accumulation mode typically account for most of the particle surface area and a considerable part of the volume (FIG. 5). PM with $d = 0.1–1$ µm exhibits the longest atmospheric residence times (Anastasio and Martin 2001), i.e. one to two weeks in the lower troposphere.

Coarse PM is generated through mechanical processes, such as abrasion and fragmentation, and includes mineral dust (e.g. from arid environments), sea salt, and biogenic and anthropogenic particles (e.g. from tire and brake abrasion). Because the volume and mass of a particle are both proportional to d^3, coarse PM typically accounts for an important fraction of the total PM volume and mass (FIG. 5). However, a larger grain size leads to a relatively short atmospheric residence time due to the high settling velocity of such particles.

Atmospheric particles display distinct size distributions in different environments: the highest concentrations are usually observed in urban areas, where the particles exhibit a number distribution dominated by ultrafine particles ($PM_{0.1}$) and a volume distribution with two distinct maxima (accumulation and coarse modes). Typical marine particles (mostly sea salt) have a prominent volume maximum in the coarse fraction, whereas remote continental particles typically have a volume maximum, albeit less pronounced, in the accumulation mode (see Fig. 5 in Grobéty et al. 2010 this issue). Remote continental particles, however, display distinctly higher number concentrations than average marine particles. These typical distributions may be modified dramatically through traffic, volcanic eruptions, dust storms, and combustion processes.

SAMPLING AND ANALYSIS

Because of the distinct physical and chemical properties of particles within the different size modes, sampling and analysis pose considerable challenges (McMurry 2000). Particles are collected by means of active or passive samplers, which use filters made of materials (e.g. cellulose, quartz, Teflon, lacey-carbon grids) chosen depending on the objective of the analysis. *Active samplers* draw ambient air, which is pumped at a specific flow rate through cutoff inlets (also known as impactors); these separate the particles according to size, so that the mode of interest accu-

mulates on the filter. *Passive samplers* collect coarse-mode particles through wet or dry deposition, i.e. through sedimentation without pumping.

Bulk chemical characterization of PM is achieved by various methods (SEE GLOSSARY). To trace the origin of particles or understand formation processes, the stable and radiogenic isotope compositions of bulk PM samples may be determined. These analytical methods, however, do not yield information on the physical, structural, and chemical characteristics of individual particles. Because such data are of great interest for deducing origin and formation processes as well as for assessing potential health impacts of particles, several single-particle methods are also in use (SEE GLOSSARY).

Sampling time needs to be adjusted depending on the objectives. For example, to collect enough PM for bulk chemical analysis, longer sampling times are used than if the goal is to collect PM for subsequent single-particle analysis. New bulk sampling methods (e.g. particle-into-liquid sampling or thermal desorption methods) can decrease considerably the sampling and analysis time for some particle components. Modern aerosol time-of-flight mass spectrometers enable the determination of inorganic and organic particle components in different submicrometer particles with a time resolution of seconds.

CHEMICAL COMPOSITION

The following categories of atmospheric PM are commonly distinguished:

- *Sea-salt particles*, which form when wind and waves force air bubbles to burst at the sea surface. They contain primarily Cl^-, Na^+, and SO_4^{2-}, with some Mg^{2+}. Interaction with nitric acid leads to the loss of Cl^- during ageing.

- *Mineral dust, volcanic ash,* and *industrial dust*, which vary widely in composition (e.g. silicates, oxides, sulfates, carbonates, alloys, glass) because of the different formation mechanisms and source compositions.

- *Primary biogenic aerosol particles (PBAP)*, which comprise biological materials (e.g. pollen, spores, plant fragments). They consist mainly of carbon, but other elements (e.g. potassium, phosphorus, silicon) may be characteristic minor components.

- *Combustion-derived carbonaceous particles*, which consist mainly of organic matter (OM) and elemental carbon (Engling and Gelencsér 2010 this issue). OM is a complex mixture of many different organic compounds with a wide range of physical and chemical properties. The
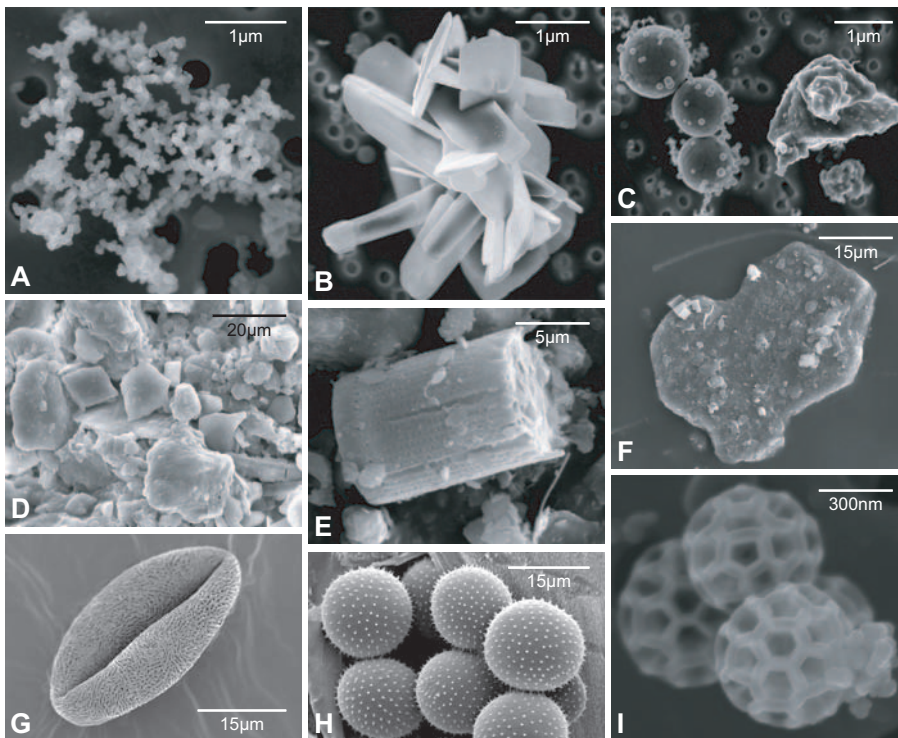
FIGURE 4 Scanning electron microscope images of anthropogenic (A–C) and natural (D–I) particles. (**A**) Typical aggregate of soot particles, Strasbourg; (**B**) combustion-derived Ca-sulfate crystals, Strasbourg; (**C**) plagioclase and spheres of combustion-derived magnetite, Strasbourg; (**D**) calcite and clays in Saharan dust, collected in Barcelona; (**E**) diatom in Saharan dust, collected at Santa Cruz de Tenerife (Canary Islands); (**F**) illite with attached halite crystals (small bright cubes) in Saharan dust, collected at Santa Cruz de Tenerife (Canary Islands); (**G**) firebush pollen (*Hamelia patens*); (**H**) spores of wheat leaf rust (*Puccinia triticina*); (**I**) agglomerate of three brochosomes (carbon-rich particles produced by leaf-hopping insects as a water-repellent body coat), Strasbourg. IMAGES COURTESY OF THE ELECTRON MICROSCOPY GROUP AT THE UNIVERSITY OF BASEL (G, H) AND THE RESEARCH GROUPS OF THE AUTHORS.

terms *elemental carbon* (EC) and *black carbon* (BC), both relatively poorly defined (SEE GLOSSARY), are used in atmospheric science for those carbonaceous materials that strongly absorb solar radiation (BC) or are highly refractory (EC). In contrast, the term *soot* is well defined (Pósfai and Buseck 2010): it refers to a primary combustion-derived particle type with a distinctive structure consisting of concentric graphene-like layers, which are wrapped into spherules that aggregate into characteristic branching clusters (FIG. 4A).

- *Secondary inorganic compounds*, which consist of salts (mainly of NO_3^-, SO_4^{2-}, and NH_4^+) that are formed from precursor gases emitted from natural (e.g. marine dimethylsulfide) or anthropogenic (e.g. SO_2, NO_x, NH_3, HCl) sources. These salts may have small contents of other cations as a result of interaction between gaseous precursors and mineral dust or sea-salt particles.

- *Secondary organic aerosol (SOA) particles*, which are composed of light elements and formed from reactive organic gases (e.g. VOCs) emitted by the biosphere (mainly forests and phytoplankton) and from anthropogenic sources.

The contents of minor and trace elements in atmospheric particles may be diagnostic of specific sources and can therefore aid in source apportionment. For example, vanadium and nickel are typically associated with the combustion of fuel oil and petroleum coke (Pacyna 1998), whereas copper, antimony, and tin in urban PM indicate that the particles are derived from abraded vehicle brake pads (Sternbeck et al. 2002). Similarly, certain organic compounds (e.g. levoglucosan) and the isotopic composition of certain elements (carbon, lead, neodymium, strontium, sulfur, zinc) can help in identifying specific sources or components in mixtures (Grobéty et al. 2010).

FLUXES

Despite large uncertainties associated with global mass-emission data, the available values (Andreae and Rosenfeld 2008; Durant et al. 2010 this issue) suggest that airborne
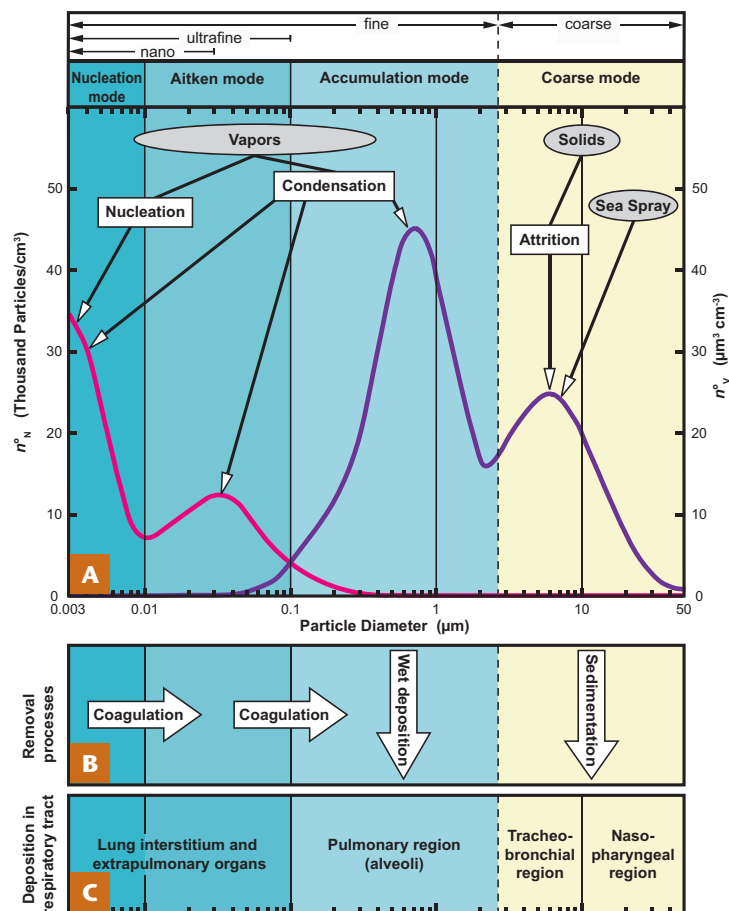

FIGURE 5 (**A**) Typical size distributions of the number (n^0_N, magenta line) and the volume (n^0_V, purple line) of atmospheric particles per cubic centimeter of air. Source materials of particles shown in grey ellipses; formation processes of particles shown in white rectangles and associated arrows. (**B**) Particle removal processes. (**C**) Areas in the respiratory tract where inhaled PM is deposited preferentially. COMPILED FROM WHITBY (1978), SEINFELD AND PANDIS (2006), AND NEWMAN (2001)
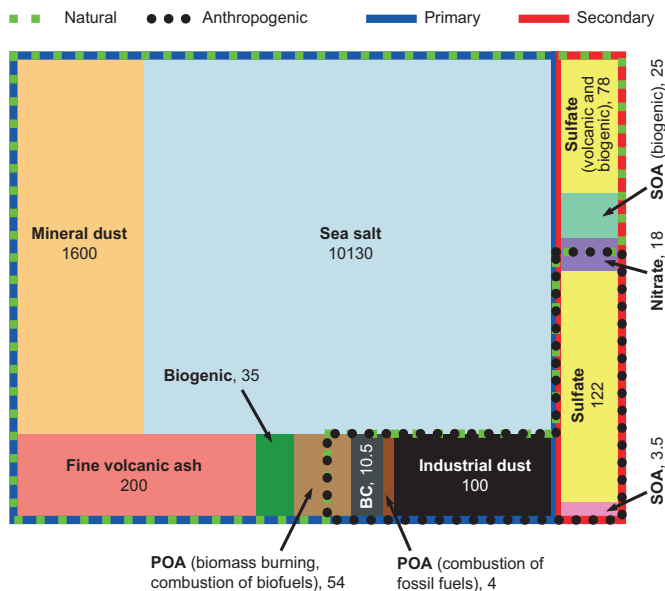
Legend: ■ ■ Natural ● ● ● Anthropogenic ▬ Primary ▬ Secondary

Mineral dust 1600

Sea salt 10130

Sulfate (volcanic and biogenic), 78

SOA (biogenic), 25

Nitrate, 18

Sulfate 122

SOA, 3.5

Biogenic, 35

Fine volcanic ash 200

BC, 10.5

Industrial dust 100

POA (biomass burning, combustion of biofuels), 54

POA (combustion of fossil fuels), 4

FIGURE 6 Fluxes of primary and secondary atmospheric PM, expressed in teragram per year (Tg = 10^{12} g = 1 Mt) and shown as a fraction of the area of a rectangle, are differentiated according to their provenance (natural and anthropogenic sources); see text. Data represent "best guess" values of Andreae and Rosenfeld (2008), except for volcanic particulate data, which are from Durant et al. (2010). Large uncertainties are associated with these estimates. For example, it is likely that biogenic SOA species are much more abundant than current estimates show. More research is needed to improve our knowledge about the contributions of the various particle types. POA = primary organic aerosol; SOA = secondary organic aerosol; BC = black carbon

particles are predominantly of natural rather than anthropogenic origin (~12,100 versus 300 Tg/y or 98 wt% versus 2 wt%; FIG. 6). Among naturally produced PM, sea salt and mineral dust (mostly from deserts) are the largest contributors to the global mass fluxes (10,130 and 1600 Tg/y, respectively). Other types of natural PM account for ~360 Tg/y (3 wt% of all natural emissions), of which fine volcanic ash (<63 µm) is the most important contributor. According to current estimates, ~99 wt% of the entire natural flux consists of primary particles, in contrast to the anthropogenic flux where secondary aerosol particles are much more prevalent (~50 wt%). This important distinction reveals that to improve air quality, it is crucial to reduce emissions of gaseous precursors, and not only emissions of primary PM. Primary anthropogenic particulates (~150 Tg/y) are generated from industrial operations, vehicle traffic (soot, abrasion particles), and combustion of biomass and fossil fuels (FIG. 6). Secondary anthropogenic particles (~140 Tg/y) are derived mostly from SO_2, NO_x, and VOCs emitted from industrial and combustion processes, as well as traffic.

These global flux estimates are not valid on a local scale, where the proportions of the different PM types may be completely different. In urban areas, for example, traffic-related PM, including resuspended dust, accounts for a much larger proportion of the total PM compared to the global estimates. Other sources, including biomass burning (wood stoves), agricultural fires, and ammonia emissions (from farming), however, contribute considerably to the PM load to which urban populations are exposed.

AIR QUALITY STANDARDS

Air quality standards are set in terms of the mass concentrations of ambient particle levels. The United States, for example, has a current daily limit value for PM_{10} of 150 µg/m³ (not to be exceeded more than once per year)

and triennial limit values for $PM_{2.5}$ of 15 µg/m³ (annual) and 35 µg/m³ (not to be exceeded on more than 7 days per year). The European Commission set the daily limit value for PM_{10} at 50 µg/m³ (not to be exceeded on more than 35 days per year). Annual values for $PM_{2.5}$ of 25 µg/m³ and 20 µg/m³ have been set as targets throughout Europe for 2010 and 2020, respectively.

The general trend followed by most air quality regulators is to focus on fine PM, because epidemiological and toxicological studies have shown that exposure to $PM_{2.5}$, which can reach the alveolar region of the lungs (FIG. 5), is linked to increases in mortality and hospital admissions due to respiratory and cardiovascular diseases (Pope et al. 2009). There is increasing evidence, however, that coarse particles may also produce adverse health effects (Brunekreef and Forsberg 2005). Coarse particles are especially abundant in areas with relatively low precipitation, high road traffic, and/or high industrial emissions. It is crucial, therefore, that fine and coarse fractions be monitored separately and that regulations exist for both modes.

Adverse health effects associated with PM exposure include chronic obstructive pulmonary disease, exacerbation of asthma, fibrosis, slower lung development in children, and lung cancer. Even though this topic is an increasingly important aspect of PM research, we will not discuss it further, as it has been reviewed in a recent issue of *Elements* (Fubini and Fenoglio 2007).

GLOBAL IMPACTS

In addition to affecting human health, PM also has profound impacts, both locally and globally, on climate, ecosystems, and visibility.

Climate

Human activities and natural processes that produce greenhouse gases, e.g. CO_2, methane (CH_4), nitrous oxide (N_2O), and halocarbons, also emit PM. Even though particles have much shorter atmospheric lifetimes than greenhouse gases, they change the radiative balance of the Earth–atmosphere system, thus contributing to climate change. These perturbations can be quantified by *radiative forcing* (RF), a term that describes how a compound or process influences the balance of incoming and outgoing energy at the tropopause (IPCC 2007). A positive RF leads to global mean surface warming, whereas a negative RF produces global mean surface cooling. The radiative effects can be categorized as direct or indirect.

Direct effects are caused by scattering and absorption of radiation by atmospheric PM, whereby dark particles (e.g. BC, soot) absorb radiation and warm the atmosphere (positive RF; FIG. 7). When deposited on snow and ice, the most reflective surfaces on Earth, dark particles lead to a considerable reduction of the reflectance, or *albedo*. Most other PM species, including sulfates and light-colored organic carbon (OC), reflect incoming solar radiation and, thus, counteract warming caused by dark particles and greenhouse gases (FIG. 7). For example, an extensive sulfate aerosol layer formed through oxidation of SO_2 injected into the stratosphere by the 1991 Mount Pinatubo eruption exerted negative RF, causing substantial cooling in the Northern Hemisphere during the summer of 1992 (Durant et al. 2010). Similarly, historic explosive volcanic eruptions caused hemispheric cooling, as documented by dendrochronological observations (Briffa et al. 1998).

Indirect effects result from the capability of hygroscopic aerosol particles to act as cloud condensation nuclei (CCN)—the seeds for condensation of water drops—or as
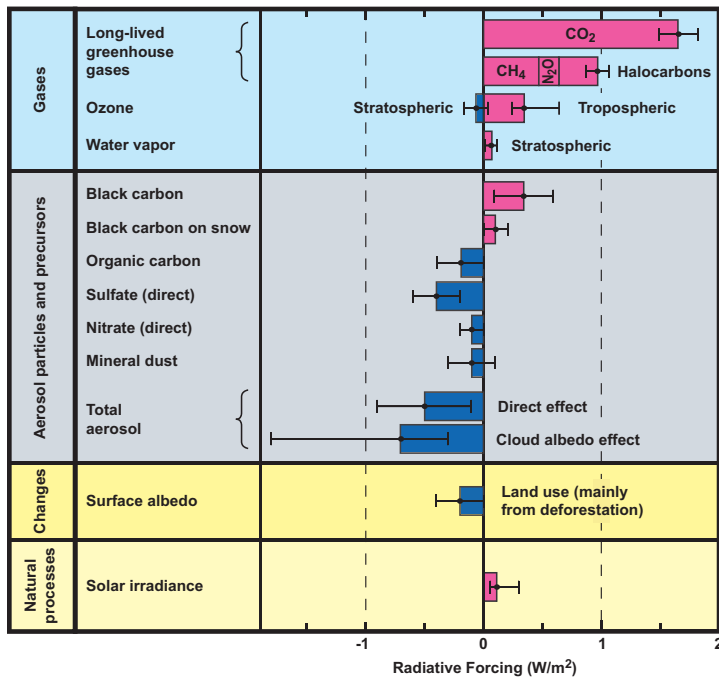
Figure 7 Summary of the radiative forcing (RF) resulting from human activities and natural processes. A positive RF leads to global mean surface warming (shown in red colors), whereas a negative RF causes global mean surface cooling (shown in blue). RF values (in W/m^2) are for changes relative to preindustrial conditions defined at 1750. Uncertainties are shown by bar lines. COMPILED FROM IPCC (2007)

aeolian dust, which also delivers other nutrients (e.g. phosphorus, silicon). Atmospheric iron deposition stimulates phytoplankton growth, since photosynthetic enzymes require iron. Enhanced phytoplankton growth, in turn, leads to an increase in the uptake of CO_2 from the atmosphere through conversion into biomass. In tropical, nitrogen-poor waters, iron can also stimulate nitrogen fixation (an energy- and iron-intensive process, which can be undertaken only by a limited number of algal species), in turn enhancing phytoplankton productivity.

However, because phytoplankton can access only the soluble fraction of iron, iron speciation in the dust particles plays a crucial role, as it determines solubility and bioavailability (Schroth et al. 2009). The speciation of iron depends not only on the mineralogical composition, and thus the source of atmospheric PM, but also on PM ageing during transport (e.g. photochemical reactions, reactions with acids, particle-size sorting; see Baker and Croot 2010). Another consequence of an iron-induced increase in marine productivity is a change in trace-gas emissions. Such a change can influence pH and hence iron availability in the water; it can also affect the composition of the atmosphere and hence its radiative properties. These biochemical and climatic effects could become more pronounced if global warming and land-use changes lead to an expansion of arid areas, thus enhancing both dust fluxes (as in the famous "Dust Bowl" of the Midwest USA) and iron supply to the surface waters of the world's oceans.

Increased dust fluxes also occurred during the Quaternary glacial intervals, when great accumulations of loess formed on several continents (Maher 2009). Increased dust deposition, however, may also have a negative effect on marine ecosystems. The observed decline of Caribbean coral reefs, for example, might be partially related to an enhanced flux of African dust, which increases the turbidity of the water. However, such environmental perturbations may also be due, in part, to the delivery of pathogens (e.g. fungal spores, bacterial cysts), which can be present in dust that has been transported over large distances, as was described by Ehrenberg in 1847.

Deposition of atmospheric PM plays a similarly important role in the biogeochemistry of soils. Both marine particles and mineral dust, derived primarily from deserts (see Engelbrecht and Derbyshire 2010 this issue), contribute significantly to the nutrients available for plant growth, especially in highly weathered, nutrient-poor soils, for example, in the Amazon rain forest and on Hawai'i (Derry and Chadwick 2007). Dust deposition, however, can also have negative impacts on soil and thus on agriculture. For example, reduction of crop yields may occur as a result of burial of seedlings, loss of plant tissue, and reduction of photosynthetic activity due to sandblasting. Lower crop yields can also be caused by deposition of PM containing heavy metals and metalloids near industrial sites.

Visibility

Major pollution episodes, volcanic eruptions, wildfires, and dust storms can greatly reduce visibility and affect, or even disrupt, transportation systems worldwide (particularly aviation), as demonstrated impressively by both historic (e.g. London's Great Smog and the U.S. Dust Bowl) and more recent episodes (the dust storm in Sydney in September 2009—see cover picture; the eruption of the volcano at Eyjafjallajökull, Iceland, in April 2010; massive forest and peat fires in Russia during July and August 2010). In some regions, such events occur on a regular basis. For example, the seasonal dust storms in eastern Asia (known as *Kosa* events or *Hwangsa*) transport substantial amounts

ice-forming nuclei (IN). The effectiveness of particles to act as CCN or IN depends on the size, chemical composition, mixing state, and number concentration of the particles. For warm clouds, an increase in PM numbers will lead to a greater abundance of smaller cloud droplets due to competition for a given water vapor content; this, in turn, increases the albedo of the cloud (*cloud albedo effect*; FIG. 7). Moreover, because smaller droplets have a larger cross-sectional area per unit mass and thus do not fall to the ground, an increase in PM numbers may prevent a cloud from raining out (see Gassó et al. 2010 this issue), thereby extending its lifetime (the *cloud lifetime effect*). Overall, this important but complex mechanism (Stevens and Feingold 2009) increases the cloud cover over Earth's surface.

The effects of PM on climate are difficult to quantify, as indicated by the large uncertainties in the RF values (FIG. 7). Much of this difficulty stems from challenges in determining the physical and chemical characteristics as well as from the spatial variability of atmospheric PM. Difficulty also results from the fact that PM undergoes significant modification while airborne (e.g. hygroscopic growth, deliquescence, reactions with acids and gases, condensation of vapors, alteration of optical properties, coagulation) (Buseck and Pósfai 1999).

Ecosystems

One of the key factors limiting phytoplankton growth over vast areas of the modern ocean is the availability of iron in surface waters, especially in *high-nutrient, low-chlorophyll* (HNLC) regions (Moore et al. 2002). Even though fluvial and glacial sediments represent the largest iron flux into the oceans, most of this iron is trapped in near-shore and shelf areas (Jickells et al. 2005). The dominant external supply of iron to the surface waters of the open ocean is

of dust from the source areas (Taklamakan and Gobi deserts, Chinese Loess Plateau) across Korea and Japan to places as far away as Greenland. Similarly, Saharan dust is observed regularly on the Canary Islands and in Europe, where it is transported by hot, seasonal winds known as *calima* and *scirocco* (or comparable winds), respectively. Another example is the southeast Asian haze episodes, which result from biomass burning and combustion of biofuels (see Engling and Gelencsér 2010 this issue).

The following articles in this issue show that geological processes and human activities taking place on one continent can have a dramatic influence on climate and ecosystems on other continents, the effects being transmitted via the atmosphere rather than the solid Earth. It is hoped that this issue of *Elements* will stimulate further research on atmospheric particles, because the effects they have are global and touch us all.

ACKNOWLEDGMENTS

REFERENCES

Anastasio C, Martin ST (2001) Atmospheric nanoparticles. Reviews in Mineralogy & Geochemistry 44: 293-349

Andreae MO, Rosenfeld D (2008) Aerosol–cloud–precipitation interactions. Part 1. The nature and sources of cloud-active aerosols. Earth-Science Reviews 89: 13-41

Baker AR, Croot PL (2010) Atmospheric and marine controls on aerosol iron solubility in seawater. Marine Chemistry 120: 4-13

Briffa KR, Jones PD, Schweingruber FH, Osborn TJ (1998) Influence of volcanic eruptions on Northern Hemisphere summer temperature over the past 600 years. Nature 393: 450-455

Brunekreef B, Forsberg B (2005) Epidemiological evidence of effects of coarse airborne particles on health. European Respiratory Journal 26: 309-318

Buseck PR, Pósfai M (1999) Airborne minerals and related aerosol particles: Effects on climate and the environment. Proceedings of the National Academy of Science 96: 3372-3379

Buseck PR, Schwartz SE (2004) Tropospheric aerosols. In: Keeling RF (Ed) Treatise on Geochemistry 4, Elsevier, New York, pp 91-142

Crutzen PJ (1976) The possible importance of CSO for the sulfate layer of the stratosphere. Geophysical Research Letters 3: 73-76

Darwin C (1846) An account of fine dust which often falls on vessels in the Atlantic Ocean. Quarterly Journal of the Geological Society 2: 26-30

Derry LA, Chadwick OA (2007) Contributions from Earth's atmosphere to soil. Elements 3: 333-338

Durant AJ, Bonadonna C, Horwell CJ (2010) Atmospheric and environmental impacts of volcanic particulates. Elements 6: 235-240

Ehrenberg CG (1847) Passatstaub und Blutregen. Abhandlungen der königlichen Akademie der Wissenschaften zu Berlin: 269-460

Engelbrecht JP, Derbyshire E (2010) Airborne mineral dust. Elements 6: 241-246

Engling G, Gelencsér A (2010) Atmospheric brown clouds: From local air pollution to climate change. Elements 6: 223-228

Fubini B, Fenoglio I (2007) Toxic potential of mineral dusts. Elements 3: 407-414

Gassó S, Grassian VH, Miller RL (2010) Interactions between mineral dust, climate, and ocean ecosystems. Elements 6: 247-252

Grobéty B, Gieré R, Dietze V, Stille P (2010) Airborne particles in the urban environment. Elements 6: 229-234

IPCC (2007) Climate Change 2007: The Physical Science Basis. Cambridge University Press, Cambridge,

Jacobson MZ (2002) Atmospheric Pollution. Cambridge University Press, Cambridge

Jickells TD and 18 coauthors (2005) Global iron connections between desert dust, ocean biogeochemistry, and climate. Science 308: 67-71

Maher BA (2009) Rain and dust: Magnetic records of climate and pollution. Elements 5: 229-234

McMurry PH (2000) A review of atmospheric aerosol measurements. Atmospheric Environment 34: 1959-1999

Moore JK, Doney SC, Glover DM, Fung IY (2002) Iron cycling and nutrient-limitation patterns in surface waters of the World Ocean. Deep Sea Research II 49: 463-507

Newman LS (2001) Clinical pulmonary toxicology. In: Sullivan JB, Krieger G (eds) Clinical Environmental Health and Exposures, Lippincott, Williams and Wilkins, pp 206-223

Pacyna JM (1998) Source inventories for atmospheric trace metals. In: Harrison RM, Van Grieken RE (eds) Atmospheric Particles. IUPAC Series on Analytical and Physical Chemistry of Environmental Systems 5, Wiley, New York, pp 387-423

Pope CA III, Ezzati M, Dockery DW (2009) Fine-particulate air pollution and life expectancy in the United States. New England Journal of Medicine 360: 376-386

Pósfai M, Buseck PR (2010) Nature and climate effects of individual tropospheric aerosol particles. Annual Review of Earth and Planetary Sciences 38: 17-43

Rottenberg D (2003) In the Kingdom of Coal. Routledge, London

Schroth AW, Crusius J, Sholkovitz ER, Bostick BC (2009) Iron solubility driven by speciation in dust sources to the ocean. Nature Geoscience 2: 337-340

Seinfeld JH, Pandis SN (2006) Atmospheric Chemistry and Physics: From Air Pollution to Climate Change. John Wiley & Sons, New York

Sternbeck J, Sjödin Å, Andréasson K (2002) Metal emissions from road traffic and the influence of resuspension—results from two tunnel studies. Atmospheric Environment 36: 4735-4744

Stevens B, Feingold G (2009) Untangling aerosol effects on clouds and precipitation in a buffered system. Nature 461: 607-613

Stone R (2002) Counting the cost of London's killer smog. Science 298: 2106-2107

Whitby KT (1978) Physical characteristics of sulfur aerosols. Atmospheric Environment 12: 135-159

Williams PI, Baltensperger U (2009) Particulate matter in the atmosphere. In: Hewitt CN, Jackson AV (eds) Atmospheric Science for Environmental Scientists. Wiley-Blackwell, Oxford, pp 168-197 ▪

Accumulation mode – A particle-size range between 100 nm and 1 µm (upper value also listed as 2 µm or 2.5 µm in the literature)

Aerosol – A dispersion of small (diameter of less than ~30 µm) solid or liquid particles suspended in a gas

Aitken mode – Particle-size range between 10 nm and 100 nm

Albedo – A measure of how strongly an object (e.g. ice) reflects radiation. It is quantified as the proportion of incident solar radiation (of all wavelengths) that is reflected.

BC – Black carbon. This collective term is used in the atmospheric sciences to describe a range of carbonaceous particles that strongly absorb shortwave solar radiation. BC is formed during combustion of fossil fuels, biofuels, and biomass and consists of various materials, including char particles, soot, and a mixture of partially burned and pyrolyzed plant materials. Most commonly, BC is determined by measuring light attenuation (e.g. by using an aethalometer)

Bulk (ensemble) methods – Methods used for bulk characterization of PM. These methods include: inductively coupled plasma mass spectrometry and optical emission spectrometry (ICP-MS and ICP-OES), aerosol mass spectrometry (AMS), X-ray fluorescence (XRF), particle-induced X-ray emission (PIXE) spectroscopy, ion chromatography (IC), thermo-optical methods for determining OC and EC contents, X-ray diffraction (XRD), and aethalometry (a method that determines the light attenuation by aerosol particles)

Cascade impactor – A device for simultaneous sampling of multiple size fractions through a series of stacked substrates, each of which collects a different particle-size range

CCN – Cloud condensation nuclei

Coarse particles – Particles with diameters of >2.5 µm. The limit of 2.5 µm was defined on a regulatory rather than scientific basis (see text). It does not coincide with the volume-distribution minimum that separates particles formed by different processes (SEE FIG. 5).

Cryosphere – A collective term describing the areas of the Earth's surface where water is present as ice. The cryosphere thus comprises ice sheets, glaciers, sea ice, lake ice, river ice, snow cover, and frozen ground (including permafrost).

Deliquescence – A process by which a solid absorbs moisture from the air until it dissolves in the absorbed water and forms a solution

Dry deposition – A term describing the overall physical process through which gases and particles are transferred directly to the surface of the Earth

EC – Elemental carbon. This is a poorly defined term, used in the atmospheric sciences to describe mostly elemental carbon in aerosol particles. EC refers to the most refractory part of carbonaceous aerosol particles, the part that oxidizes above a certain threshold value (established in combustion experiments). It is determined by thermal or thermo-optical methods.

Fine particles – Particles with diameters of <2.5 µm ($PM_{2.5}$). The limit of 2.5 µm was defined on a regulatory rather than scientific basis (see text). It does not coincide with the volume-distribution minimum that separates particles formed by different processes (SEE FIG. 5).

Fine volcanic ash – Volcanic ash particles with diameters of <63 µm

HULIS – Humic-like substances. These chemical compounds are similar to the humic and fulvic acids in soils, but form primarily during biomass burning and also during secondary chemical reactions in the atmosphere (i.e. particle ageing).

Hydrometeor – An atmospheric particle formed by condensation of water vapor or freezing of water

Hygroscopicity – A term describing the tendency of a substance to absorb moisture from the air (but not necessarily to the point of dissolution)

Impactor – A device for separating PM according to size so that the size mode of interest accumulates on a sampling substrate

IN – Ice-forming nuclei

Mixing state – A term describing whether aerosol particles occur as single or multiple phases. Single-phase particles are known as *externally mixed*, whereas multiple-phase particles are called *internally mixed*.

Nucleation mode – A particle-size range with diameters of <10 nm

OC – Organic carbon. The term describes the mass of carbon contained in all organic compounds present in an aerosol particle. OC is typically determined by thermo-optical methods, along with EC.

OM – Organic matter. This is a collective term describing thousands of individual organic compounds, which vary widely in regard to physical and chemical properties.

PAH – Polycyclic aromatic hydrocarbon. PAHs are aromatic compounds (e.g. anthracene, benzo[a]pyrene, naphthalene), some of which are carcinogenic and mutagenic. They are produced during combustion of fossil fuels and biomass burning.

PAN – Peroxyacetyl nitrates. These substances form photochemically from nitrogen dioxide (NO_2) and hydrocarbons emitted into the atmosphere by combustion processes. They are respiratory and eye irritants.

PBAP – Primary biogenic aerosol particles (e.g. plant debris, animal fragments, pollen, spores, algae, fungi, bacteria, and viruses)

PCB – Polychlorinated biphenyl. PCBs are toxic organic compounds with 1–10 chlorine atoms attached to biphenyl, an aromatic hydrocarbon.

PM (PM_{10}, $PM_{2.5}$, $PM_{0.1}$) – Particulate matter (with diameters of <10 µm, <2.5 µm, and <0.1 µm, respectively)

POA – Primary organic aerosol particles. These are particles emitted directly into the atmosphere

RF – Radiative forcing. A term describing how a compound or process influences the balance of incoming and outgoing energy at the tropopause. A positive RF leads to global mean surface warming, whereas a negative RF produces global mean surface cooling.

Single-particle methods – Methods used to characterize individual aerosol particles. These methods include: scanning electron microscopy (SEM) combined with energy-dispersive X-ray (EDX) spectroscopy, electron probe microanalysis (EPMA), transmission electron microscopy (TEM) combined with selected-area electron diffraction (SAED), particle-induced X-ray emission (µ-PIXE) spectroscopy, confocal laser-scanning microscopy (CLSM), optical microscopy combined with automated image analysis, synchrotron-based X-ray microscopy (e.g. NEXAFS, µ-XRF), aerosol time-of-flight mass spectrometry (ATOFMS).

SOA – Secondary organic aerosol particles. Particles formed in the atmosphere from volatile organic compounds (VOCs).

Soot – A primary combustion-derived particle type with a characteristic and well-defined structure consisting of graphene-like layers, which are wrapped into spherules with diameters of 20–50 nm. These spherules aggregate into clusters that typically have a chain-like fractal appearance (FIG. 4A). Soot strongly absorbs solar radiation and reemits it as heat, thus exerting a strong positive RF.

Ultrafine particles – Particles with diameters of <0.1 µm ($PM_{0.1}$)

VOC – Volatile organic compound. These organic substances have relatively high vapor pressures and are of both natural (e.g. terpene) and anthropogenic (e.g. formaldehyde, acetone) origin.

Wet deposition – A term describing the overall physical process through which rain removes materials from the atmosphere. Wet deposition can be further subdivided into "rain-out" (in-cloud scavenging) and "wash-out" (below-cloud scavenging).