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Atmospheric Aerosols: Chemical Reactions and Effects on Climate

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ABSTRACT: Atmospheric aerosols consist of small particles of solids, like dust, and liquids, like water, suspended in the atmosphere. Atmospheric aerosols can be either emitted directly into the atmosphere as a particle, like ash, or form when emitted gases undergo complex chemical reactions and condense as particles. Aerosols also have a wide range of sources and can be further classified as natural or anthropogenic, which originate from human-related activities. There are many different types of natural aerosols, including dust, sea-salt, smoke, and those from living sources, referred to as biogenic aerosols.

Atmospheric aerosols range in size from a few nanometers—the size of a DNA molecule—to tens of microns—the width of a human hair—in diameter. The size of a particle alters its behavior, including how long it remains suspended in the air. Aerosol systems are complex, undergoing chemical reactions that can cause the aerosols to evaporate and recondense. This complexity makes it challenging to predict and model aerosol properties.

KEYWORDS: atmospheric, aerosols, particles, chemical reactions

I. INTRODUCTION

Atmospheric aerosols play a significant role in Earth's climate. Depending on their composition, they may either absorb or reflect heat and sunlight. They also form necessary seeds for cloud formation. In many climate models, the uncertainty around atmospheric aerosols and their interaction with clouds remains one of the largest overall sources of uncertainty in climate projections.

Atmospheric aerosols affect air quality and have consequences for human health. Directly measuring the amount and type of aerosols in a specific region can provide insight into how factors like pollution and wildfires influence air quality. Measurements of aerosol types can help researchers identify the sources of unknown aerosols and determine how they form

Pacific Northwest National Laboratory (PNNL) scientists are actively working toward developing a comprehensive understanding of atmospheric aerosols. Their research includes creating and improving aerosol models, measuring the atmosphere to determine how aerosols behave in different areas of the globe, and mimicking aerosol formation, growth, and evaporation processes in a controlled laboratory environment.

More accurately representing atmospheric aerosols is important for modeling applications ranging from determining how thunderstorms behave to assessing the Earth's energy budget in a changing climate. PNNL researchers have active roles in developing and implementing aerosol models for projects over a range of scales. Using PNNL's expertise in modeling Earth systems, scientists are working to better model aerosols and aerosol-cloud interactions in Earth system models through the Energy Exascale Earth System Model and Enabling Aerosol-cloud interactions at GLOBal convection-permitting scales (EAGLES) projects. PNNL researchers have also worked to enhance aerosol representation in regional models, like the Weather Research and Forecasting Model (WRF).[1,2,3]

PNNL staff are deeply involved in field campaigns through the U.S. Department of Energy's Atmospheric Radiation Measurement (ARM) user facility. In these campaigns, researchers use ground- and aircraft-based equipment to collect atmospheric data, including the amount and type of aerosols present. Data from the Amazon, the U.S. Great Plains, and the Azores, for example, have helped identify the sources of different types of aerosols. This research provides an important baseline because the Amazon and Azores are relatively isolated locations with minimal human-influenced aerosols, that can be compared to measurements made in more polluted locations.

Atmospheric aerosol research at PNNL benefits from the Atmospheric Measurements Laboratory (AML) located on the Richland campus. The AML has an atmospheric research chamber that enables scientists to simulate atmospheric

conditions in a controlled environment. This facilitates studies of how volatile organic gases form aerosol particles and how those aerosols change over time. The AML also contains an ice nucleation chamber capable of mimicking how aerosols become seeds for clouds. These experimental capabilities provide data that lead to enhanced understanding of aerosol processes.

The combination of these three classes of expertise and capabilities allows PNNL researchers to work on large, multifaceted projects. The Integrated Cloud, Land-Surface, and Aerosol System Study (ICLASS) focuses on understanding and modeling interactions between clouds, aerosols, and land. Such an undertaking requires data from field campaigns, facilities for laboratory experiments, and expertise in developing and analyzing models.

II. DISCUSSION

All atmospheric aerosols scatter incoming solar radiation, and a few aerosol types can also absorb solar radiation. BC is the most important of the latter, but mineral dust and some OC components are also sunlight absorbers. Aerosols that mainly scatter solar radiation have a cooling effect, by enhancing the total reflected solar radiation from the Earth. Strongly absorbing aerosols have a warming effect. In the atmosphere, there is a mixture of scattering and absorbing aerosols, and their net effect on Earth's energy budget is dependent on surface and cloud characteristics. Scattering aerosols above a dark surface and absorbing aerosols above a bright surface are most efficient (see Figure 3a). Scattering (absorbing) aerosol above a bright (dark) surface are less efficient because the solar radiation is reflected (absorbed) anyway. Absorbing aerosols are particularly efficient when positioned above clouds,[4,5,6] which are a main contributor to the total reflection of solar radiation back to space.

Aerosols are vital for cloud formation because a subset of them may serve as cloud condensation nuclei (CCN) and ice nuclei (IN). An increased amount of aerosols may increase the CCN number concentration and lead to more, but smaller, cloud droplets for fixed liquid water content. This increases the albedo of the cloud, resulting in enhanced reflection and a cooling effect, termed the cloud albedo effect (Twomey 1977; Figure 3b). Smaller drops require longer growth times to reach sizes at which they easily fall as precipitation. This effect, called the cloud lifetime effect, may enhance the cloud cover (see illustration in Figure 3b) and thus impose an additional cooling effect (Albrecht 1989). However, the life cycles of clouds are controlled by an intimate interplay between meteorology and aerosol-and-cloud microphysics, including complex feedback processes, and it has proven difficult to identify the traditional lifetime effect put forth by Albrecht (1989) in observational data sets.

Absorbing aerosols also have the potential to modify clouds properties, without directly acting as CCN and IN, by: (1) heating the air surrounding them while reducing the amount of solar radiation reaching the ground, which stabilizes the atmosphere and diminishes the convection and thus the potential for cloud formation, (2) increasing the atmospheric temperature, which reduces the relative humidity, inhibits cloud formation, and enhances evaporation of existing clouds. This is collectively termed the semi-direct aerosol effect (Hansen et al. 1997). The net effect is uncertain (see Figure 3b) and highly depends on the vertical profile of BC (Koch & Del Genio 2010).

In addition, BC and other absorbing aerosols deposited on snow or ice surfaces may reduce the surface albedo, leading to reduced reflectance of solar radiation, and hence a heating effect (Hansen & Nazarenko 2004).

Radiative forcing (RF) is often used to quantify and compare the potential climate impact of the various aerosol effects. RF is defined as a change[7,8,9] in the Earth's radiation balance due to a perturbation of anthropogenic or natural origin.. The total aerosol forcing probability density function (PDF), in addition to individual aerosol components, indicating both the magnitudes and uncertainty of the effects, is shown in Figure 4a. The wider a PDF, the larger is the uncertainty. Combining all aerosol effects (blue dashed curve in Figure 4a) enhances the uncertainty compared to considering only the direct aerosol effect and cloud albedo effect. Here c is the heat capacity of the land-ocean-atmosphere system and λ is the climate sensitivity. At radiative equilibrium ($d(\Delta T)/dt = 0$), Equation 1 reduces to $\Delta T = \lambda RF$. However, the Earth is not in radiative equilibrium, since less thermal radiation is currently emitted to space compared to what is absorbed of solar radiation (Hansen et al. 2005). This radiative imbalance causes the Earth to gradually warm, with global warming as a result (Trenberth & Fasullo 2010). The simple equation above has two key uncertainties. The observed surface temperature change is rather well determined over the industrial era, but the climate sensitivity and the total RF are both highly uncertain. The climate sensitivity is an essential parameter for prediction of future climate change. Quantifying the climate sensitivity for the doubling of CO₂ has long been attempted by using global climate models or temperature records, but it still has a wide range of reported values (IPCC 2007, Knutti & Hegerl 2008). The total RF through the industrial era is also uncertain, mainly due to lack of quantification of the

aerosol effects discussed above. The implication of this uncertainty in the aerosol RF for the quantification of the climate sensitivity can be illustrated as follows:

If we assume a total aerosol RF and a current energy imbalance, we can compute the resulting climate sensitivity using Equation 1 (Figure 4b). This can then be compared with the PDFs for the current aerosol RF to get an indication of the range in climate sensitivities allowed by the present knowledge (red and blue lines in figure 4b). A similar figure has previously been presented in Andreae et al. (2005). [10,11,12] The allowed climate sensitivity ranges from about 2 to 8 Kelvin (K) for a doubling of CO₂ using the known industrial age warming of around 0.8 K, the present best knowledge of RF from non-aerosol components, the 90% confidence interval of the total aerosol RF for the most certain effects, and radiative imbalance.

III. RESULTS

There has been a tremendous improvement in the understanding of atmospheric aerosols and their climate effect over the last decades, with some important observational and modelling breakthroughs. Long-term measurements of aerosols (e.g., Putaud et al. 2010, Andrews et al. 2011), observational campaigns (e.g., Quinn & Bates 2005), and remote sensing from space and ground (Holben et al. 1998, Remer et al. 2008) have remarkably increased knowledge about the composition and characteristics of atmospheric aerosols. However, an understanding of the greater complexity of atmospheric aerosols has at the same time limited more robust quantification of their climate effect. The first estimate of the direct aerosol effect in the early 1990s was limited to sulphate aerosols (Charlson et al. 1991), with estimates for BC coming a few years later (Haywood & Shine 1995). Observations have shown that OC is an important aerosol component (Novakov et al. 1997, Ramanathan et al. 2001), and substantial investigations have later explored the complex composition and optical characteristics of this compound (e.g., Kanakidou et al. 2005, Graber & Rudich 2006). Global aerosol models today provide RF estimates for a large set of aerosol components, such as sulphate, BC (from fossil fuel and biomass burning), OC (primary and secondary from fossil fuel and biomass burning), and nitrate (Jacobson 2001, Liao & Seinfeld 2005, Koch et al. 2009). In addition, multi-model studies are performed to understand and reduce uncertainties due to model differences (Schulz et al. 2006).

An example of recent progress is reduced uncertainty in the estimate of the total direct aerosol effect. This estimate was made possible by advances that have occurred on both the modelling and the observational side, and was based on a combination of global aerosol models and observation based methods (mostly remotely sensed data). [13,14,15] Initially, observational estimates of RF were up to three times stronger than model based calculations (Forster et al. 2007). Consistency between these two different approaches has subsequently been reached, and was found to arise from necessary and simplified assumptions of the pre-industrial aerosol composition in the observation-based method (Myhre 2009). Although the uncertainty in the total direct aerosol effect is reduced, it is still substantial compared to uncertainties associated with greenhouse gases. In addition the uncertainty in individual RF for several of the aerosol components, such as BC, OC, and nitrate, is large.

Similar to the early estimates of the direct aerosol effect, many of the first model estimates of the aerosol indirect effect only accounted for the effect of sulphate particles acting as CCN (Kaufman & Chou 1993, Jones et al. 1994). Furthermore, they only included the influence of sulphate aerosols on cloud albedo, disregarding any effects on cloud lifetime and extent. With the realization that other aerosol species of anthropogenic origin could also form cloud droplets and that effects on cloud lifetime and extent were also possible, global climate models estimated the aerosol indirect effect to be stronger (e.g., Lohmann & Feichter 1997, Menon et al. 2002). Some even predicted this cooling effect to be comparable in magnitude to the warming greenhouse effect. Recent publications have later pointed to oversimplifications in model representation of clouds and how their lifetimes are affected by aerosols (e.g., Stevens & Feingold 2009). It is now acknowledged that aerosol effects on cloud lifetime will vary with the cloud type in question, and that complex feedback processes can sometimes complicate the ultimate cloud response to aerosol perturbations. Recent model studies have found that by forming ice in super-cooled liquid clouds, aerosols may in fact shorten cloud lifetime, because of the more efficient precipitation formation when cloud ice is present (e.g., Lohmann & Hoose 2009, Storelvmo et al. 2011). In summary, whether aerosols are acting as CCN or IN or are simply modifying atmospheric stability by absorbing solar radiation, there is still high uncertainty associated with their effect on cloud lifetime. This uncertainty reflects how challenging it is to represent aerosol-and-cloud processes that occur on microscopic scales in models that have resolutions of tens to hundreds of kilometres. Although much uncertainty remains, model and satellite estimates [16,17,18] of the cloud albedo effect seem to converge on a negative RF that has about half the magnitude of the positive RF attributed to increasing CO₂ concentrations.

Several types of atmospheric aerosol have a significant effect on Earth's climate: volcanic, desert dust, sea-salt, that originating from biogenic sources and human-made. Volcanic aerosol forms in the stratosphere after an eruption as droplets of sulfuric acid that can prevail for up to two years, and reflect sunlight, lowering temperature. Desert dust, mineral particles blown to high altitudes, absorb heat and may be responsible for inhibiting storm cloud formation. Human-made sulfate aerosols, primarily from burning oil and coal, affect the behavior of clouds.^[17]

Although all hydrometeors, solid and liquid, can be described as aerosols, a distinction is commonly made between such dispersions (i.e. clouds) containing activated drops and crystals, and aerosol particles. The atmosphere of Earth contains aerosols of various types and concentrations, including quantities of:

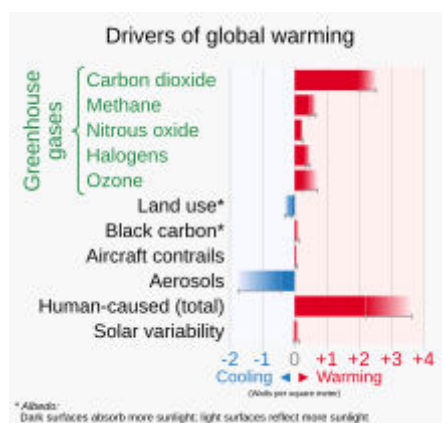
- natural inorganic materials: fine dust, sea salt, or water droplets
- natural organic materials: smoke, pollen, spores, or bacteria
- anthropogenic products of combustion such as: smoke, ashes or dusts

Aerosols can be found in urban ecosystems in various forms, for example:

- Dust
- Cigarette smoke
- Mist from aerosol spray cans
- Soot or fumes in car exhaust

The presence of aerosols in the Earth's atmosphere can influence its climate, as well as human health.

Effects



Aerosols have a cooling effect that is small compared to the radiative forcing (warming effect) of greenhouse gases.^[18]

Volcanic eruptions release large amounts of sulphuric acid, hydrogen sulfide and hydrochloric acid into the atmosphere. These gases represent aerosols and eventually return to earth as acid rain, having a number of adverse effects on the environment and human life.^[19]

When aerosols absorb pollutants, it facilitates the deposition of pollutants to the surface of the earth as well as to bodies of water.^[20] This has the potential to be damaging to both the environment and human health.

Aerosols interact with the Earth's energy budget in two ways, directly and indirectly.

- E.g., a direct effect is that aerosols scatter and absorb incoming solar radiation.^[21] This will mainly lead to a cooling of the surface (solar radiation is scattered back to space) but may also contribute to a warming of the surface (caused by the absorption of incoming solar energy).^[22] This will be an additional element to the greenhouse effect and therefore contributing to the global climate change.^[20]
- The indirect effects refer to the aerosol interfering with formations that interact directly with radiation. For example, they are able to modify the size of the cloud particles in the lower atmosphere, thereby changing the way clouds reflect and absorb light and therefore modifying the Earth's energy budget.^[19]
- There is evidence to suggest that anthropogenic aerosols actually offset the effects of greenhouse gases in some areas, which is why the Northern Hemisphere shows slower surface warming than the Southern Hemisphere,

although that just means that the Northern Hemisphere will absorb the heat later through ocean currents bringing warmer waters from the South.^[23] On a global scale however, aerosol cooling decreases greenhouse-gases-induced heating without offsetting it completely.^[24]

Aerosols in the 20 μm range show a particularly long persistence time in air conditioned rooms due to their "jet rider" behaviour (move with air jets, gravitationally fall out in slowly moving air);^[25] as this aerosol size is most effectively adsorbed in the human nose,^[26] the primordial infection site in COVID-19, such aerosols may contribute to the pandemic.^[27]

Aerosol particles with an effective diameter smaller than 10 μm can enter the bronchi, while the ones with an effective diameter smaller than 2.5 μm can enter as far as the gas exchange region in the lungs,^[28] which can be hazardous to human health.^[19]

IV. CONCLUSION

Particles can deposit in the nose, mouth, pharynx and larynx (the head airways region), deeper within the respiratory tract (from the trachea to the terminal bronchioles), or in the alveolar region.^[57] The location of deposition of aerosol particles within the respiratory system strongly determines the health effects of exposure to such aerosols.^[57] This phenomenon led people to invent aerosol samplers that select a subset of the aerosol particles that reach certain parts of the respiratory system.^[58]

Examples of these subsets of the particle-size distribution of an aerosol, important in occupational health, include the inhalable, thoracic, and respirable fractions. The fraction that can enter each part of the respiratory system depends on the deposition of particles in the upper parts of the airway.^[59] The inhalable fraction of particles, defined as the proportion of particles originally in the air that can enter the nose or mouth, depends on external wind speed and direction and on the particle-size distribution by aerodynamic diameter.^[60] The thoracic fraction is the proportion of the particles in ambient aerosol that can reach the thorax or chest region.^[61] The respirable fraction is the proportion of particles in the air that can reach the alveolar region.^[62] To measure the respirable fraction of particles in air, a pre-collector is used with a sampling filter. The pre-collector excludes particles as the airways remove particles from inhaled air. The sampling filter collects the particles for measurement. It is common to use cyclonic separation for the pre-collector, but other techniques include impactors, horizontal elutriators, and large pore membrane filters.^[63]

Two alternative size-selective criteria, often used in atmospheric monitoring, are PM10 and PM2.5. PM10 is defined by ISO as particles which pass through a size-selective inlet with a 50% efficiency cut-off at 10 μm aerodynamic diameter and PM2.5 as particles which pass through a size-selective inlet with a 50% efficiency cut-off at 2.5 μm aerodynamic diameter. PM10 corresponds to the "thoracic convention" as defined in ISO 7708:1995, Clause 6; PM2.5 corresponds to the "high-risk respirable convention" as defined in ISO 7708:1995, 7.1.^[64] The United States Environmental Protection Agency replaced the older standards for particulate matter based on Total Suspended Particulate with another standard based on PM10 in 1987^[65] and then introduced standards for PM2.5 (also known as fine particulate matter) in 1997^[20]

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