A Contribution to the Encyclopedia of Climate and Weather

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Aerosols - Aerosols are fine solid or liquid particles suspended in the atmosphere, where they reside typically for days to weeks before settling to the ground or being washed out by rain or snow. They arise both from natural sources (such as desert dust, sea spray and volcanic eruptions) and from human activities involving burning of fossil fuels and vegetation. Visible forms of atmospheric aerosol plumes include dust, smoke, smog and haze. Aerosol particles are small, but numerous, and often comprise of a number of inorganic and organic substances. They adversely affect human health, and thus are regulated as air pollutants. As an integral component of the Earth's climate system, aerosols interact strongly with sunlight, and provide condensation nuclei for cloud formation. Man-made aerosols alter the climate in fundamental ways.

History of Study Modern aerosol science finds its root in the so-called Age of Enlightenment, during which many hypotheses on the origin of aerosols were postulated. Scattered observations made in the 19th century helped rule out some of them (such as earthquakes, thunderstorm lightning and meteoric dust). In 1783, Benjamin Franklin, then the United States ambassador to France, speculated that a volcanic eruption in Iceland may have caused the hazy condition over the Western Europe, and the blocking of sunlight by haze may be responsible for that year's unusually cold summer. In 1888, J. Kiessling showed convincingly that the 1883 explosion of the Krakatau volcano (called Krakatoa at the time) in the East Indies injected large quantities of aerosols into the atmosphere, which dimmed the sun around the world for months.

John Aitken pioneered quantitative measurements of atmospheric aerosols in the 1880s. His dust counter worked by expanding and cooling a sample of air containing suspended particles. Water vapor then condensed and grew on each particle to form droplets. The small cloud created in this way was projected against a grid, and the droplets were counted using a small microscope. Important work on the growth of water droplets on dust particles and on ions was carried out in a classic investigation by C. T. R. Wilson in 1897. This work led to the invention of cloud chamber, a device for detecting the tracks of charged elementary particles. Much of the early work on aerosols and ions was done at the Cavendish Laboratory in Cambridge, England, at the turn of the 20th century.

Aerosol science started to emerge as an independent discipline during the Second World War. Research performed as part of the classified Manhattan Project formed the basis of the field's first handbook. In the few decades that followed, the concerns over public health prompted researchers to study the fallout (radioactive dust) from nuclear weapon tests and the worsening photochemical smogs (a toxic "cocktail" of particulate matter and gas-phase pollutants) in densely populated cities (such as London and Los Angeles).
In the 1970s, a small group of scientists made the earliest attempts to quantify the climate impacts of anthropogenic aerosols with rudimentary mathematical models. They did not agree upon whether aerosols warm or cool the Earth, let alone the effect's magnitude. Since then, as it has become increasingly evident that locally emitted aerosols and

precursors have global reach by being transported over long distances by atmospheric circulation, more concerted efforts, experimental and theoretical alike, have been made to better understand the physical and chemical processes that are key to determining their climate impacts. Aerosols are now routinely measured within the atmosphere with a variety of sophisticated instruments aboard aircrafts, and from space with passive and active sensors aboard satellites. The measurements are used to constrain the representation of aerosols in global climate models and to evaluate the realism of model simulations. The current consensus is that the net aerosol effect is one of cooling, which has "masked" a substantial portion of the global warming caused by the green-house gases. Yet, the magnitudes of different aerosol effects, particularly those arising from cloud changes, are still uncertain.

Emission sources Both natural and anthropogenic emissions contribute to the formation of atmospheric aerosols. Another useful distinction is between primary and secondary aerosols. The former are emitted directly into the atmosphere, while the latter are generated thorough chemical reactions within. Naturally occurring primary aerosols are commonly created by mechanical disruption, or cleaving. They include wind-blown dust from newly plowed fields or deserts, and crystals of sea salt formed through evaporating sea sprays generated by breaking waves over the oceans. Some relatively large particles are biological in nature (such as pollen grains, spores, and fragments of leaves).

An important source of man-made primary aerosols is widespread biomass burning, especially in the tropics. Farmers burn dry vegetation seasonally to prepare land for shifting cultivation and to re-fertilize the soil. In developing countries, biomass burning is an energy source for cooking, heating, and charcoal production. Burning is also a means of deforestation. Naturally occurring and accidental wildfires are also important sources of primary aerosols in the boreal regions. Aerosols generated from biomass burning and wildfires consist mostly of black carbon (soot) and organic carbon.

The formation of secondary aerosols inside a parent gas phase requires gas-to-particle conversion, which is often realized through nucleation. Generally speaking, what is necessary for nucleation to take place is an overabundance or supersaturation of one or more chemical species. An important type of secondary aerosols produced by nucleation is sulfuric acid. Powerful volcanic eruptions can inject gigantic amounts of sulfur dioxide gas directly into the stratosphere, where it is oxidized into sulfuric acid vapor. In the presence of relatively abundant water vapor, even infinitesimal amounts of sulfuric acid vapor can nucleate into nascent particles.

Human-induced burning of fossil fuels (coal, oil and natural gas) containing tracer amounts of sulfur is the main source of tropospheric sulfate aerosols. Sulfuric acid from burning coal also causes acid rain. Natural emission sources participate in the tropospheric sulfur cycle as well. An important oceanic source is the microscopic phytoplankton in sea water, which produce a sulfur-bearing substance called dimethyl sulfide (DMS). This organic gas, which humans identify with "the odor of the sea", is believed to be a major source of the sulfuric acid particles at the rare pristine locations unaffected by industrial air pollution.

Photochemical reactions of volatile organic compounds (VOC) can produce secondary organic aerosols (SOA). For example, terpenes (a class of natural VOC) emitted from conifer trees can form particles that are composed of organic species and a few hundreds of a micron in diameter. These SOA particles are thought to be responsible for the blue haze over the Smoky Mountains in Tennessee, which was reported before the beginning of industrial activity in the region. VOC and nitrogen oxides released from automobile exhaust, when undergoing photochemical reactions, generate SOA and tropospheric ozone, two harmful "ingredients" of urban smogs.

Long-range Transport Aerosols are light, and remain in the atmosphere long enough to be moved by winds to places far away from where they were originally emitted. As such a transport knows no boundary, it is often a source of international disputes. Sir Francis Bacon reported in ca. 1600 probably the first such incident. The Gasgogners in southern France filed a complaint with the King of England claming that their wine crop was seriously damaged by the smoke from the springtime burning of seaweed in Sussex, England.

As revealed vividly by satellite images, the springtime dust plumes from the Gobi and Taklamakan deserts in Mongolia and China can be carried by strong westerly winds across the Pacific, and reaches North America in a matter of days. This atmospheric "conveyor belt" also carries man-made pollutants including sulfate aerosols from East Asia, and has a negative impact on air quality in the western United States. In a similar fashion, the United States "exports" pollution to Europe, and European pollution finds it ways to the northern Africa and Near East. The easterly trade winds in the Tropics blow the Saharan dust across the Atlantic to the Caribbean and southwest United States. This trans-boundary behavior of air pollution makes international cooperation necessary for devising effective control strategies.

Size Distribution Individual aerosol particles are members of what is termed a population. A way of looking at the population distribution of particles is to speak of the amount of aerosol mass in different size classes. Even though there are many more small particles than large ones, the former may not comprise more mass than the latter. This way of looking at aerosol populations in terms of the amount of aerosol mass in different size intervals proves useful as new technology becomes available for classifying aerosols accurately by size. Both nature and industrial processes produce aerosol size distributions shaped like the well-known Gaussian probability distribution called the log-normal distribution. This provides scientists with a simple mathematical tool to use in building models for calculating aerosol effect as, for example, in climate models.

Three modes of aerosol mass are common in the atmosphere. First, we frequently find a mode of very small particles in the atmosphere, of the order of 0.1 to 0.01 micron in diameter. This mode is produced by chemical reactions among atmospheric trace gases. A second persistent mode, almost always observed, is composed of removal-resistant particles in the atmosphere. These are about 0.1 to 1 micron in diameter. This group is sometimes called the accumulation mode. The so-called large or giant particles are grouped into the third mode. This population consists of spores, pollen, and other biological material, as well as minerals broken off by erosion and weathering.

Chemical and Physical Transformation After aerosols are formed, they continue to be modified in the atmosphere. Some of the modifications are chemical in nature. Sulfuric acid, for instance, is neutralized into ammonium sulfate or ammonium bisulfate by ammonia gas arising from decaying biological matter. Sea salt particles become coated with waxy hydrophobic (water repellent) surface layers. The sharp edges of broken crystalline material are softened by condensed acid films. The final aerosol product sampled from the atmosphere is sometimes so modified that it bears little chemical or physical resemblance to the source materials.

An important physical process involves the collision and coagulation of different particles. Even when the particles are of the same chemical composition, coagulation complicates the aerosol structure, frequently changing its surface area and sometimes giving rise to new compounds from chemical reactions among the collisional byproducts. Very small particles like those produced by gas-to-particle conversion are quite mobile, zigzagging violently in the air as air molecules collide with them. The so-called Brownian motion of these particles makes them tend to diffuse to other larger particles and build up surface layers on them. Thus, crystals of salt or clay may soon become covered with a liquid film of sulfuric acid through constant collision with the tiny droplets of sulfuric acid in suspension. This “wets” the crystal’s surface and alters its chemistry, especially when particles act as condensation nuclei for the formation of cloud droplets.

The chemical compositions of man-made aerosols vary with industrial processes. Aerosols from automobile exhaust were rich in lead and bromine before unleaded gasoline came into use. Petroleum refineries use vanadium as a catalyst, and a tiny fraction of the metal makes its way into the produced combustion oil. Coal often contains elevated amounts of manganese, while smelter smoke from nonferrous metals is rich in arsenic, selenium, and indium. Research is in progress to relate the chemical components of aerosols to regional sources as a method of tracking air pollution.

Biogeochemistry Phytoplankton are ubiquitous in the oceans. They absorb large amounts of atmospheric CO_2 through photosynthesis, constituting an important link in the Earth’s carbon cycle. The growth of phytoplankton is often limited by the availability of soluble iron, a necessary nutrient. Deposited dust particles are the main suppliers of iron over the open oceans, and thus have an important role in determining the abundance of phytoplankton and oceanic uptake of CO_2. Higher iron input from dust could lower atmospheric concentration of CO_2 by “fertilizing” the oceans. However, it is still unclear how dust emission and transport, both of which are sensitive to the meteorological variables like winds and rainfall, would vary in a changing climate.

Radiative Properties A major fraction of atmospheric aerosols are of a size close to the wavelengths of visible light. Thus, they can reflect sunlight back to space efficiently, and tend to cool the Earth. Pure inorganic aerosols (such as sulfate and sea salt) are entirely scattering. When the ambient air is humid, they swell by taking up moisture. This so-called hygroscopic growth enhances scattering as it increases aerosol mass. On the other hand, the fact that certain types of aerosols have colors means that they can absorb visible light to some degrees. Black carbon from incomplete combustion of biomass and fossil fuels is a very good absorber, while organic carbon and dust are mildly absorbing. When a purely scattering (transparent) particle becomes internally mixed with an absorbing (colored) particle, the combined particle as a whole is absorbing (colored). This change in mixing state tends to enhance the overall absorption.

Interestingly, whether a layer of absorbing aerosols warms or cools the Earth depends on how bright or dark the underlying surface is. Smoke flowing out over a bright snow-covered surface is likely to appear dark, while the same smoke flowing over a dark surface (such as a forest) may appear lighter in color. In the first case, a net heating of the atmosphere-surface system is introduced by the smoke's additional light absorption, while in the second, the region below the smoke would cool slightly. Atmospheric aerosols from pollution sources generally contain a small fraction of black carbon. Therefore, they can either warm or cool a region, depending on the surface albedo or reflectance. Aerosols flowing over the bright ice- and snow-covered white surface in the polar regions introduce a heating effect, while they force a slight cooling in the mid-latitudes.

Cloud Formation Aitken's and Wilson's early studies showed that the tiny water droplets in clouds form on aerosols. Not all aerosols are equally efficient in promoting droplet formation. In the 1920s, Hilding Kohler carried out important work on the continuous growth of aerosols into cloud droplets, which provided the theoretical framework for understanding the roles of aerosols in cloud formation. Much of this fundamental work underpins today's research on aerosol-cloud interactions. Kohler's theory indicated that it is more difficult for cloud droplets to form on insoluble aerosols, say dust, than on soluble ones, say sulfate and sea salt. Water vapor also nucleates more readily on larger aerosols than on smaller ones. This explains why the relatively large and soluble members of the aerosol population are preferred as cloud condensation nuclei (CCN).

In the years following the Second World War, serious attempts were made to modify the amount and type of precipitation from clouds by dispersing man-made CCN in the atmosphere. These cloud seeding projects stimulated a lot of work on cloud physics. In the l940s, W. E. Howell calculated cloud activation on aerosols. These complex calculations required digital computers, then in their infancy. A great step forward came in the late 1950s when an Irish physicist, Sean Twomey, developed useful mathematical formulae to describe the cloud activation process. Twomey's work showed that the number of cloud droplets depends strongly on the size distribution and solubility of the aerosol population, and that an increase in the concentration of aerosols often resulted in an increase in that of cloud droplets. If the amount of cloud water is unchanged, more numerous droplets lead to a reduction in the average droplet size, and an increase in the droplet surface area available for scattering sunlight. As a result, the clouds become brighter. This linkage between aerosols and cloud albedo is on vivid display in the satellite images of “ship tracks”, the brightening of marine boundary layer clouds caused by the aerosol particles in ship exhaust.

Climate Effects Aerosols influence the Earth’s climate mainly in two ways. When the sky is clear (devoid of clouds), aerosols can reflect incoming sunlight back to outer space. This blocks part of the energy that would have reached the surface, and acts to cool the climate. Absorbing aerosols, black carbon in particular, are able to trap solar energy within the atmosphere. Though absorption, like reflection, tends to reduce sunlight at the ground level, the enhanced atmospheric heating eventually warms up the surface, and counteracts the cooling caused by reflection. These are the direct climate effects of aerosols.

Both Twomey’s theoretical work and observations suggest that cloud reflectance or albedo goes up with the number of aerosol particles that provide nucleating sites for forming cloud droplets. The resulting brightening of clouds renders them more potent at shielding the surface from sunlight, giving rise to the so-called first indirect effect (or cloud albedo effect). More aerosols may also enable clouds to last longer by suppressing rainfall. This is the second indirect effect (or cloud lifetime effect).

As a regular occurrence in the atmosphere, clouds are important for regulating the climate. A cooling of 3°C would result from increasing cloud reflectance by about 5 percent. Roughly speaking, this degree of cooling would be sufficient to offset the global warming from doubling the atmospheric concentration of carbon dioxide. This is why anthropogenic aerosols can have a substantial cooling effect on the climate by altering clouds, and may have “masked” a significant fraction of the anthropogenic greenhouse gases-induced warming. However, it is by no means certain that we can count on aerosol-induced cooling to balance greenhouse warming since aerosols have a much shorter lifetime in the atmosphere than carbon dioxide. Besides, the current estimates of aerosol effects are still subject to large uncertainties.

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