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**PERSPECTIVES**

**ATMOSPHERIC SCIENCE:**

# Reshaping the Theory of Cloud Formation

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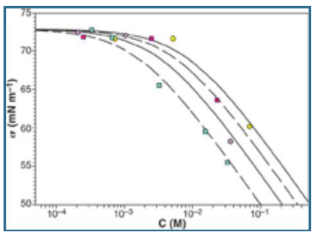
Droplet clouds are the most important factor controlling the albedo (reflectivity) and hence the temperature of our planet. Man-made aerosols have a strong influence on cloud albedo, with a global mean forcing estimated to be of the same order (but opposite in sign) as that of greenhouse gases (1), but the uncertainties associated with the aerosol forcing are large. Recent studies indicate that both the forcing and its magnitude may be even larger than anticipated.

Cloud optical properties are controlled by the sizes and numbers of the droplets in the cloud, which are, in turn, governed by the availability of atmospheric particles that serve as cloud condensation nuclei. Twomey (2) suggested that an increase in atmospheric aerosols from anthropogenic emissions would lead to smaller cloud droplets because the same amount of cloud liquid water is distributed among more condensation nuclei. For the same liquid water content, a cloud with more numerous, but smaller, drops has a higher albedo than one with fewer, larger drops. This phenomenon, termed the first indirect climatic effect of aerosols, could constitute a major climate forcing (1). But current estimates of indirect aerosol radiative forcing or of its uncertainty (1) do not include the combined influences of some recently identified chemical factors, each of which leads to additional negative forcing (cooling) on top of that currently estimated.

Estimates of the indirect climatic effect of aerosols are based on the theory of cloud droplet formation advanced by the Swedish scientist Hilding Köhler in the 1920s and 1930s (3, 4). Köhler assumed that clouds consist of "activated" water droplets that grow spontaneously after they have reached a critical size corresponding to a critical value of the supersaturation of water vapor. Köhler further assumed that the aerosol is composed of a completely soluble salt and that the particles are in thermodynamic equilibrium until the point of spontaneous growth. Indeed, it is still generally assumed that a cloud forms only in a supersaturated water environment with all the solute coming from the particle. It has recently become clear, however, that soluble gases (5, 6), slightly soluble solutes (7), and surface tension depression by organic substances (8) also influence the formation of cloud droplets, in a manner unforeseen by Köhler.

Nitric acid ($HNO_3$) is perhaps the most important highly soluble trace gas in the atmosphere. Ample data establish the prevalence of nitrate as a constituent of cloud and fog water in polluted air (9-11). In the presence of a water-soluble trace gas such as $HNO_3$, the critical supersaturation for that droplet is lowered as the gas condenses into a growing droplet. Depending on how it is dispersed over the aerosol population, a minute amount of soluble gas can exert a profound effect on the number of activated droplets. A striking consequence of the presence of a soluble trace gas is that clouds or fogs with micrometer-sized droplets may exist even though the droplets have not undergone traditional activation and even though the ambient relative humidity never exceeds 100% (5, 6). Such "pollution clouds" have a higher droplet number concentration and a broader droplet size distribution than "clean clouds" (12).

Highly soluble gases are not the only compounds that can affect aerosol activation. The importance of carbonaceous compounds as components of atmospheric aerosols is well established. A variety of measurements have shown that between 20 and 60% of the carbon mass in fine (diameter <1 μm) atmospheric aerosols consists of partially soluble organic compounds (13-16). A partially soluble aerosol component adds solute to the aqueous phase as the droplet grows, decreasing the critical supersaturation of the particle. Many of these organic compounds are surface active (see the figure) (8); if, in addition, surface tension is lowered as the substance dissolves, the critical supersaturation is further lowered, and the number of particles that can activate increases even more. In general, the lowering of surface tension associated with a dissolving substance has a stronger effect on cloud properties than the fact that the substance itself is only partially soluble, given that most water-soluble organic compounds are surface active.




**Surface tension lowering by organics in cloud water.** Surface tension decrease with respect to pure water as a result of water-soluble organic carbon in cloud water (expressed as moles per liter of carbon). Data from Tenerife (Spain) and Po Valley (Italy) taken by one of the authors (M.C.F.).

As predicted by Köhler some 80 years ago, droplet activation places an upper limit on the supersaturation of water vapor that can be reached in the atmosphere. Given sufficient solute or enough depression of surface tension, or a combination of the two, the supersaturation in a given situation will decrease. At high aerosol and soluble trace gas concentrations and for low cooling rates, strict activation is not necessary for formation of a visible cloud; indeed, a continuum exists from ambient aerosol to wetter and wetter particles to unactivated clouds to activated ones. What is seen as "cloud" can, in reality, be a collection of droplets ranging from fully activated to unactivated.

By affecting cloud optical properties, these chemical phenomena may lead to nonnegligible global negative forcing (17) and may be as important regionally as the Twomey effect itself. To assess the importance of the indirect climatic effect of aerosols, one seeks a robust connection between cloud droplet population and a prognostic variable from global aerosol models. How that link might depend upon chemical cloud activation effects, including variations in aerosol chemical composition, solute water solubility, solute surface tension lowering, and condensation of trace gases, remains to be determined. Lack of global data on these activation effects poses additional uncertainty beyond that already

recognized by the Intergovernmental Panel on Climate Change (1), making the largest uncertainty in estimating climate forcing even larger.

**References and Notes**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17. Preliminary simulations of cloud albedo differences relative to a baseline case, in which the cloud is formed from an aerosol composed exclusively of soluble salt, show that the presence of surface tension-lowering substances or a soluble gas ($HNO_3$) leads to an increase in the number of activated droplets and cloud albedo that, if applied globally, is of a climatological significance.


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