questions. The formation of complex A requires one tyrosine in the first cycle of the enzyme. But once complex A is formed, the iron site at which this chemistry takes place is at least partially blocked by CO and CN and is electronically very different from that in the resting state of HydG. It remains to be shown how tyrosine is subsequently converted to CO to form complex B (together with a surplus CN^-) in the second cycle of HydG, and how the $Fe(CO)_2(CN)$ synthon is transferred and coupled to give the dithiolate-bridged subsite in the H-cluster.

By beginning to explain the early stages of H-cluster biosynthesis, the elegant spectroscopic study by Kuchenreuther *et al.* extends our knowledge of how the metallo-sulfur active sites of a range of enzymes are assembled from simpler building blocks. The accessory proteins HydE, HydF, and HydG are all involved in the in vivo assembly of the active H-cluster on HydA (*7*) and must play a role in some or all of these steps. However, Kuchenreuther *et al.* have previously shown in vitro that HydG without HydE or HydF can activate an apo-hydrogenase in the presence of reductant and lysate (*10*). Given that in the native pathway, all the iron in the subsite moiety originates from HydG (*5*), it is plausible that a diiron subsite forms from two (or possibly one) HydG synthons, with subsequent release into solution and capture by apo-HydA1. Support for this idea comes from a study showing that a synthetic diiron subsite (see the figure) (*11*) can be captured from solution by apo-HydA1 to give an active FeFe hydrogenase without participation of HydE or HydF (*12*).

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10.1126/science.1249276

ATMOSPHERIC SCIENCE

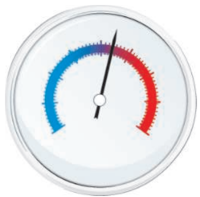
Climate Effects of Aerosol-Cloud Interactions

Advances in satellite observations and model development are needed to disentangle the complex interactions of aerosols and clouds and their effects on climate.

Daniel Rosenfeld[1]*, Steven Sherwood[2], Robert Wood[3], Leo Donner[4]

Aerosols counteract part of the warming effects of greenhouse gases, mostly by increasing the amount of sunlight reflected back to space. However, the ways in which aerosols affect climate through their interaction with clouds are complex and incompletely captured by climate models. As a result, the radiative forcing (that is, the perturbation to Earth's energy budget) caused by human activities is highly uncertain, making it difficult to predict the extent of global warming (*1*, *2*). Recent advances have led to a more detailed understanding of aerosol-cloud interactions and their effects on climate, but further progress is hampered by limited observational capabilities and coarse-resolution climate models.

Recent advances have revealed a much more complicated picture of aerosol-cloud interactions (see the figure) than considered previously. For example, radiative forcing due to aerosol-cloud interactions may be limited by buffering mechanisms that result in compensation between different cloud responses to aerosols (*3*). Other situations may be hypersensitive to aerosols because aerosols have become extremely depleted by precipitation (*4*). In these ultraclean regimes, addition of aerosols can dramatically increase cloud cover, causing a large cooling (*5*). Another newly appreciated process is aerosol-induced invigoration of deep convective clouds that may transport larger quantities of smaller ice particles to the anvils of such clouds. The higher, colder, and more expansive anvils can lead to warming by emitting less thermal radiation to space (*6*).



Challenges in CLIMATE SCIENCE

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The Intergovernmental Panel on Climate Change's fifth assessment report (*2*) begins to account for some of these aerosol cloud–mediated effects. Most studies address a subset of known or suspected mechanisms, and they generally cannot separate individual contributions. Yet, this represents advancement with respect to the fourth assessment report (*7*), which accounted for only one specific effect: the aerosol-induced reduction of cloud drop size and the resultant increasing cloud solar reflectance. It is now clear that the reduced cloud drop size triggers other processes that may induce larger radiative perturbations than the droplet-size effect through mechanisms such as those depicted in the figure (*8*). The inability to fully quantify these effects increases the uncertainty in the radiative forcing of aerosols and clouds. Furthermore, little is known about the unperturbed aerosol level that existed in the preindustrial era. This reference level is very important for estimating the radiative forcing from aerosols (*9*). Quantification of the reference level requires better quantitative understanding of the natural and anthropogenic emission sources and their interactions.

At fine scales (tens of meters or less), the processes by which aerosols alter the formation and growth of cloud drops and by which drops coalesce into rain are comparatively well understood, as are the ways in which turbulence affects these processes. Less clear is the response of the cloud cover and organization to the loss of water by rainfall. Understanding of the formation of ice and its interactions with liquid droplets is even more limited, mainly due to poor ability to measure the ice-nucleating activity of aerosols and the subsequent ice-forming processes in clouds. Explicit computer simulations of these processes even at the scale of a whole cloud or multicloud system, let alone that of the planet, require hundreds of hours on the most powerful computers available. Modelers must therefore resort to simple parametric representations of these processes

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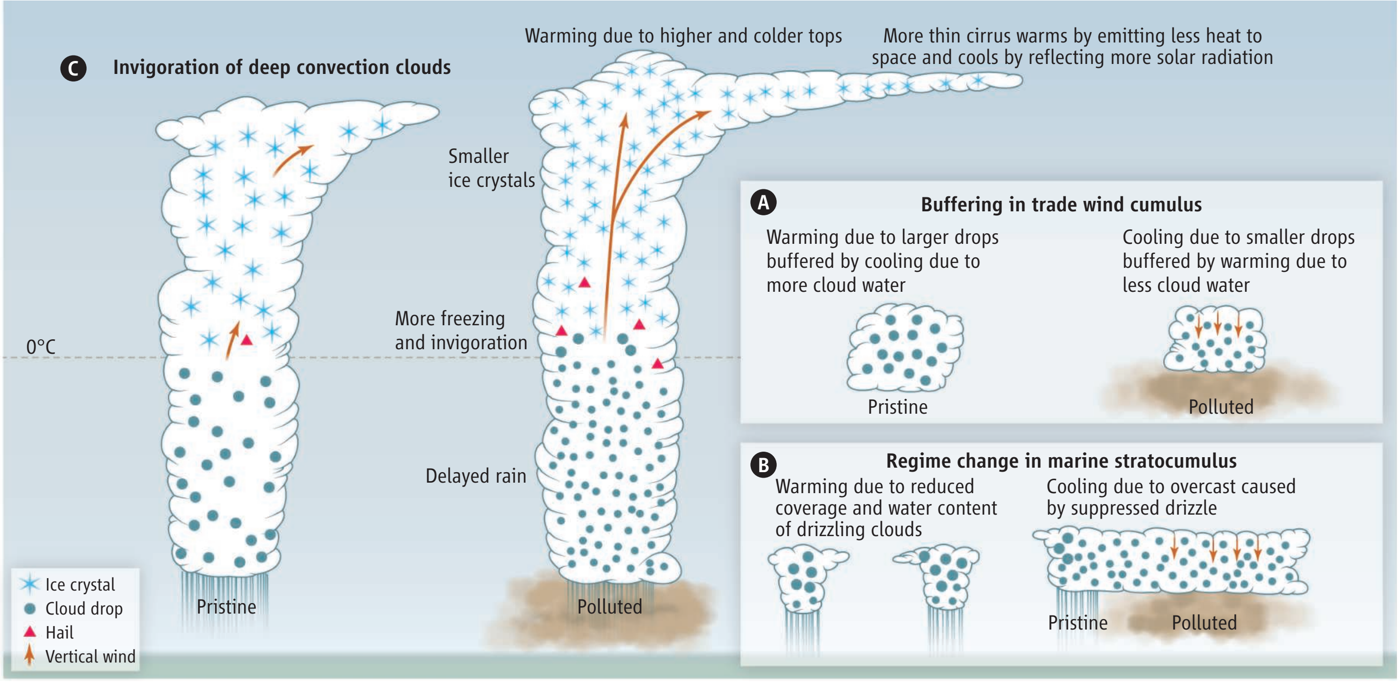

How aerosols affect the radiative properties of clouds. By nucleating a larger number of smaller cloud drops, aerosols affect cloud radiative forcing in various ways. (**A**) Buffering in nonprecipitating clouds. The smaller drops evaporate faster and cause more mixing of ambient air into the cloud top, which further enhances evaporation. (**B**) Strong cooling. Pristine cloud cover breaks up by losing water to rain that further cleanses the air in a positive feedback loop. Aerosols suppressing precipitation prevent the breakup. (**C**) Larger and longer-lasting cirrus clouds. By delaying precipitation, aerosols can invigorate deep convective clouds and cause colder cloud tops that emit less thermal radiation. The smaller ice particles induced by the pollution aerosols precipitate more slowly from the anvils. This can cause larger and longer-lasting cirrus clouds, with opposite effects in the thermal and solar radiation. The net effect depends on the relative magnitudes.

in weather and climate models. Representing aerosol effects on clouds is particularly challenging, because small-scale correlated variations between aerosol and cloud properties have large-scale consequences, such as changes in cloud organization.

Fully resolved, global, multiyear simulations are not likely to become feasible for many decades. However, an exciting step was made in recent groundbreaking simulations (*10*), in which small domains capable of resolving cloud-scale processes, including simplified schemes of cloud aerosol interactions, were embedded in each grid cell of a climate model. This approach offers the potential for model runs that resolve clouds on a global scale for time scales up to several years, but climate simulations on a scale of a century are still not feasible. The embedded model is also too coarse to resolve many of the fundamental aerosol cloud processes at the scales on which they occur.

Improved observational tests are essential for validating the results of simulations and ensuring that modeling developments are on the right track. Current satellites can measure cloud and precipitation properties but not the vertical winds that create the clouds, nor the specific aerosols on which the cloud drops and ice crystals nucleate. Therefore, it is difficult to disentangle the aerosol and meteorological effects on cloud properties. A major challenge is that the most important aerosol nucleation region is at the bottom of a cloud, which is obscured by the rest of the cloud if measured from above.

The Cloud-Aerosol Lidar and Infrared Pathfinder Satellite Observation (CALIPSO) (*11*) and EarthCare satellite missions aim to address some of these challenges. EarthCare (estimated to launch in 2015) will measure vertical profiles of aerosol types and amounts with a 355-nm lidar. EarthCare's Doppler cloud radar can determine cloud vertical motions. However, the radar and lidar in both missions cover only the line of subsatellite track, limiting their coverage. Further satellite missions that overcome the measurement challenges are being considered (*12*).

Progress on understanding aerosol-cloud interactions and their effects on climate is limited by inadequate observational tools and models (*13*). Yet, achieving the required improvement in observations and simulations is within our technological reach. For example, available technology could provide multispectral and multiangle polarimetric measurements of cloud properties at a resolution of 100 m (*12*). The level of effort should match the socioeconomic importance of what the results could provide: lower uncertainty in anthropogenic climate forcing and better understanding and predictions of future impacts of aerosols on our weather and climate.

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Acknowledgments: D.R. is partially supported by project BACCHUS FP7-603445. The authors appreciate V. Ramaswamy's comments on this Perspective.

10.1126/science.1247490

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