The Cloudy Science of Aerosols

Seeking the Unknown Quantity in Climate-Change Predictions

Models of climate change will fail to provide accurate predictions unless they account for the impact of aerosol particles and the clouds that form around them. Los Alamos researchers have taken to the skies to quantify the effects of aerosols and create better cloud models.

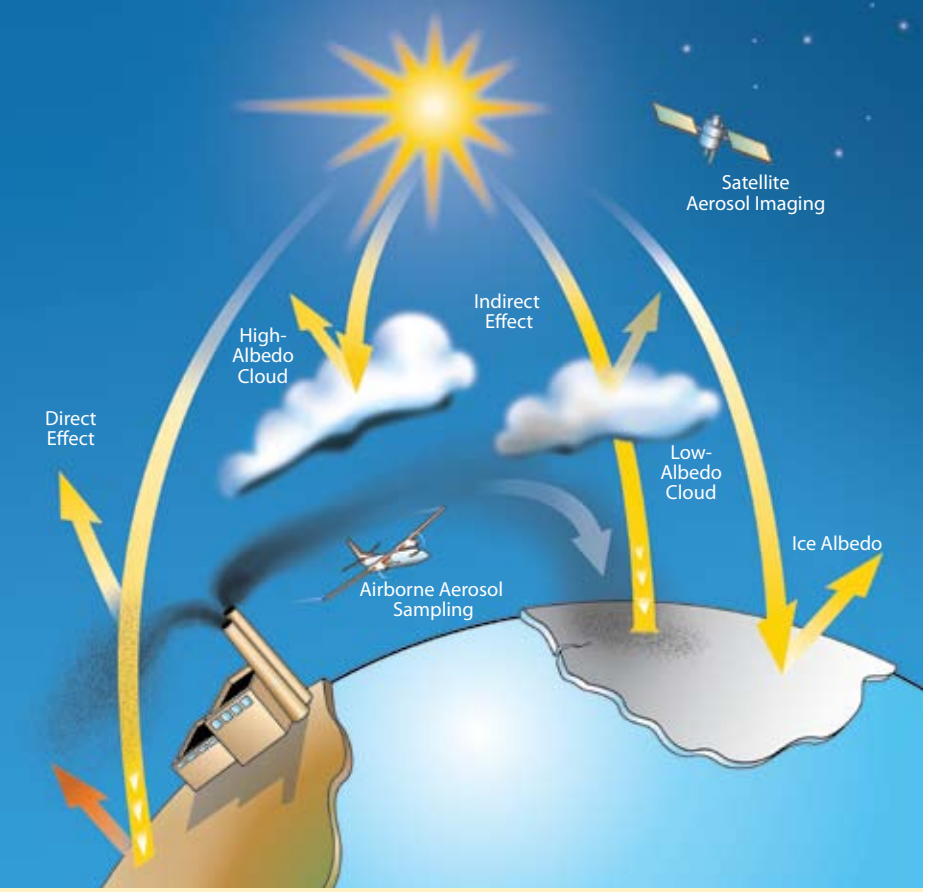
Every child, watching white fleecy shapes shift in the sky, naturally wonders where clouds come from. Most will probably learn that clouds form when water evaporates from lakes, rivers, and oceans and condenses into droplets that eventually fall as rain. Though mostly correct, that explanation is missing an important piece. Cloud droplets need airborne particles, called "aerosols," around which to condense. These particles are hurled into the sky naturally from dust storms, volcanoes, sea spray, and fires, but many come from human industrial and agricultural activities. That's right; those graceful, pristine puffs of cotton candy are also made of pollution and dirt that humans and the planet spew into the atmosphere.

Aerosols are tiny, ranging from 1/1000th of a micron to 100s of microns—anywhere between the size of a virus and a grain of sand. The period at the end of this sentence, at about 500 microns, would be an extremely large aerosol. Though miniscule, aerosols have an enormous effect on global climate by directly reflecting or absorbing solar radiation. Aerosols also indirectly affect the climate because they influence the size and abundance of cloud droplets, which in turn determine how much sunlight a cloud reflects and how much rain it produces.

Human-generated aerosols alter global climate, as do their better-known stepsisters, the greenhouse gases, but aerosols' effects are more complicated—so complicated that the Intergovernmental Panel on Climate Change (IPCC) declared them the greatest source of uncertainty in predicting climate change. And that's a big problem.

Just as a Florida homeowner relies on the local weather forecast to know if it's time to board up the windows, policy makers around the world need long-term, global-climate predictions to

Left: A wing-mounted probe collects aerosol samples over Alaska. Above: Inside the aircraft, Manvendra Dubey analyzes the samples using a photoacoustic instrument.



Climatic Effects of Aerosols

Aerosols, tiny particles suspended in the air, affect the Earth's energy balance by absorbing or reflecting solar radiation. This is called the aerosol direct effect. Aerosols also have an indirect effect by influencing cloud properties, such as lifetime, height, and the number and size of droplets. Clouds with fewer but larger droplets reflect less solar radiation (low albedo) while those with high aerosol concentrations reflect up to 90 percent of radiation back into space (high albedo). Dark aerosols from industry can settle on Arctic ice, which normally reflects sunlight, and accelerate melting by absorbing sunlight. Los Alamos researchers are analyzing aerosols using airborne and satellite techniques.

plan appropriate responses. Current climate models predict that by the end of this century there will be a temperature increase of 1.2ºC to 4.4ºC—a range far too wide to be a useful prediction. Aerosols are the unknown quantity.

To reduce the uncertainty, climate scientists must do two things: first, identify a precise numerical value for the effect aerosols have on the planet's energy budget. The Earth maintains a balance between incoming solar radiation and amounts reflected back into space or absorbed and re-radiated as infrared. Changes in the balance are known as "radiative forcing." Earth's preindustrial equilibrium is the baseline, and scientists need to accurately measure changes to it caused by human-generated aerosols. Second, scientists must gain a better understanding of the interactions between aerosols and clouds to accurately represent them in predictive models. To achieve these goals, researchers at Los Alamos National Laboratory are using the Laboratory's unparalleled computing power and advanced tools for gathering and interpreting atmospheric data.

Climate's Gray Area

We know that greenhouse gases, like CO_2, warm the planet, and we even know how much, but what about aerosols? "When you burn fossil fuels," explains Manvendra Dubey of Los Alamos' Earth and Environmental Sciences Division, "you emit not only CO_2, but also sulfur dioxide (SO_2), which becomes sulfate aerosols. Sulfates, along with some other aerosols, cool the planet by reflecting sunlight away." In the 1970s and 80s, before smokestacks were equipped to scrub sulfur, industry produced so much sulfate pollution that the resultant cooling counteracted the warming of greenhouse gases. But health concerns over particulate pollution and acid rain brought about the Clean Air Act, which forced industry to reduce sulfate emissions and ironically allowed the greenhouse effect to intensify. "Because we succeeded in reducing sulfate pollution," says Dubey, "we must now work twice as hard to control CO_2."

So if, as we learned with the sulfates, aerosols cool the planet, all we need to do is figure out how much and plug that number into climate models, right? Unfortunately, the problem is not so black and white—literally. Some aerosols, like sulfates and sea salt particles, may cool because they're white and reflect sunlight, but other aerosols, such as black carbon (soot), are dark and absorb sunlight like a black shirt on a hot summer day. When they all mix into one giant atmospheric pointillist painting, you get shades of gray whose effects are hard to quantify. But scientists are working to measure the amount of light reflected and absorbed by the aerosol mix—what they call "aerosol optical depth" (AOD). AOD is the key parameter in determining the elusive radiative-forcing number needed for precise climate predictions.

Soot and other aerosol pollutants rise into the atmosphere above Mexico City.
PHOTO BY NANCY MARLEY, ARGONNE NATIONAL LABORATORY

Listening to Dust

Conventional methods of measuring AOD can exaggerate the darkness by a factor of 2 or 3, but Dubey has corrected that error by deploying the world's first aircraft-mounted, three-laser, photoacoustic instrument. As a plane flies through clouds and haze, the instrument sucks in particles and exposes them to light from red, green, and blue lasers that together represent the solar spectrum. When particles absorb the laser energy and heat up, they expand the air around them and create a sound wave that is detected by a highly sensitive microphone. Sensors also detect light reflected by the particles. The two measurements together translate into an accurate measurement of AOD.

Dubey has flown over Mexico City, Korea, Houston, California, and even the Arctic to collect and analyze aerosol mixtures with the laser instrument. In Mexico City, he measured the effects of megacity pollution on global warming. In addition to finding sulfates and other kinds of aerosols he anticipated, he also detected a significant amount of aerosols produced by organic gases that vehicles emit. When the sun rises, these gas molecules undergo a photochemical reaction that turns them into particles dubbed "secondary organic aerosols." They were considered to be negligible, but climate modelers are now including them in chemical-transport simulations.

In Jeju, an island off the coast of South Korea and downwind from China, Dubey analyzed aerosols blowing in from Beijing to see if China had taken effective steps to clean the air for the 2008 Olympics. The Chinese government would not allow soot measurements within their country, but wind ignores borders, so Jeju was the next best thing.

But perhaps the most interesting of Dubey's observations were those obtained in the Arctic because they threw a new twist into the aerosol story. "You expect the Arctic to be pristine," says Dubey, "but it's pretty polluted." And the pollution isn't from local particles, as in Mexico City, but from an international mix of junk from all over the Northern Hemisphere. Dubey observed plumes similar to Los Angeles smog coming from Siberian fires, Gobi Desert dust storms, and industrial emissions. The new twist in the story is that the gray mix of aerosols, which contains a lot of black carbon, affects not just the atmosphere but also the ice itself. The aerosols settle on the Arctic ice sheet, causing it to absorb solar radiation and melt faster than computer models have predicted. Clean Arctic ice normally has a cooling, "albedo" effect, reflecting solar radiation.

A closer look at radiative forcing helps illustrate the significance of this phenomenon. Radiative forcing is expressed in watts per square meter (W/m^2). If a climatic influence warms the planet, as greenhouse gases do, it causes positive forcing. If it cools, as do sulfate aerosols, it causes negative forcing. Over the last century, human-caused greenhouse gases have produced a positive radiative forcing of 2.6 W/m^2 while aerosols are estimated to have had a negative radiative forcing of –1.2 W/m^2, though that figure is still highly uncertain. These measurements are global, long-term averages that don't take regional and seasonal effects into account. Those are the very effects that current ice-melt models lack. During the period Dubey studied the Arctic, the radiative forcing of black carbon for that region was a whopping 30 W/m^2.

"It's a double whammy," says Dubey. "Black carbon takes away the negative forcing of ice albedo and adds positive forcing directly to the ice surface." And it couldn't happen in a worse place.

In some places, regional and temporary disturbances might not impact global climate, but the Arctic isn't one of those places. Petr Chylek, a pioneer in aerosol science and a frequent collaborator with Dubey, explains why: "If global warming occurs, what disaster awaits humankind? Temperatures rose 0.7°C over the last 125 years, but you can't feel it. If it goes up another degree here, nothing happens. The danger is in the Arctic because melting Arctic ice results in rising sea levels. If the Greenland ice sheet melts, we have a global disaster."

Watching Aerosols from Space

Chylek was thinking about how aerosols affected climate long before it was a hot topic. Thirty-four years ago, while a researcher at the National Center for Atmospheric Research, Chylek published a paper entitled "Aerosols and Climate" in the prestigious journal *Science*. In that 1974 paper, he pointed out the need for accurate aerosol measurements and ended on a note of hope, speculating that "someday their effect may be measured directly when changes in the albedo of the earth-atmosphere system are remotely monitored

Petr Chylek

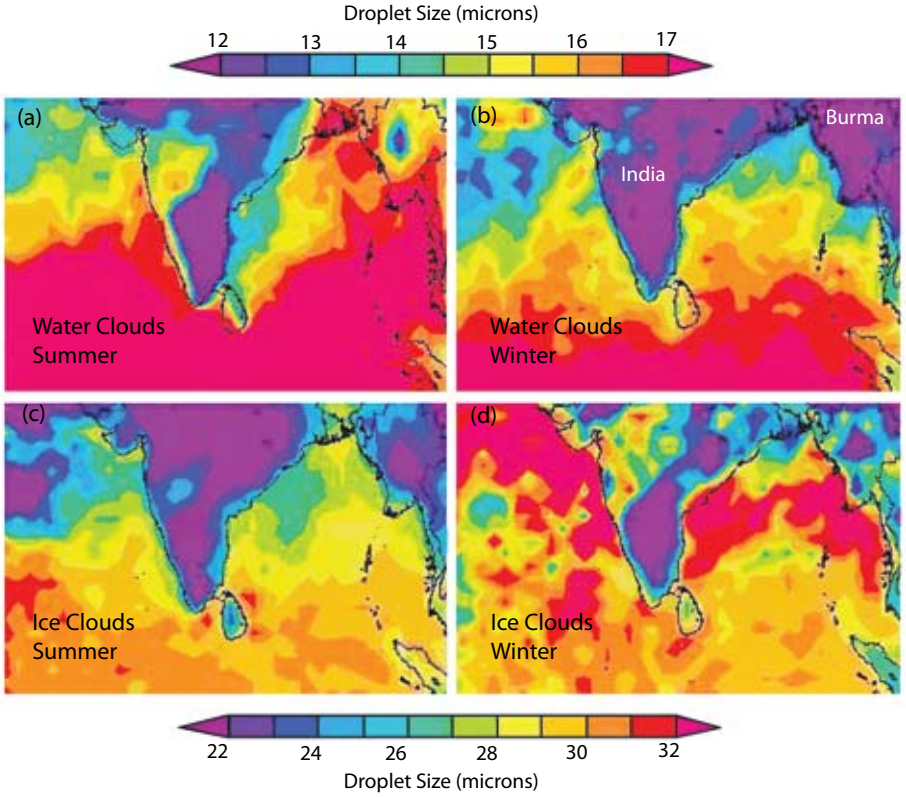
by satellites." That day has come and Chylek, now remote-sensing team leader in Los Alamos' International, Space, and Response Division, is now in the satellite business.

Chylek's work builds on Los Alamos' history of using satellites to detect the illicit production of weapons of mass destruction. One of those satellites, the Multispectral Thermal Imager (MTI), has circled the planet since March 2000, collecting images of the Earth with instruments that see changes in light and heat that the human eye cannot. While looking for the telltale gases, dusts, and heat produced by chemical or nuclear activity, the MTI has produced mountains of environmental data. Chylek took on the task of figuring out what nondefense questions might also be answered with MTI's data, and there was the answer to his hopes from 1974—global pictures of aerosols.

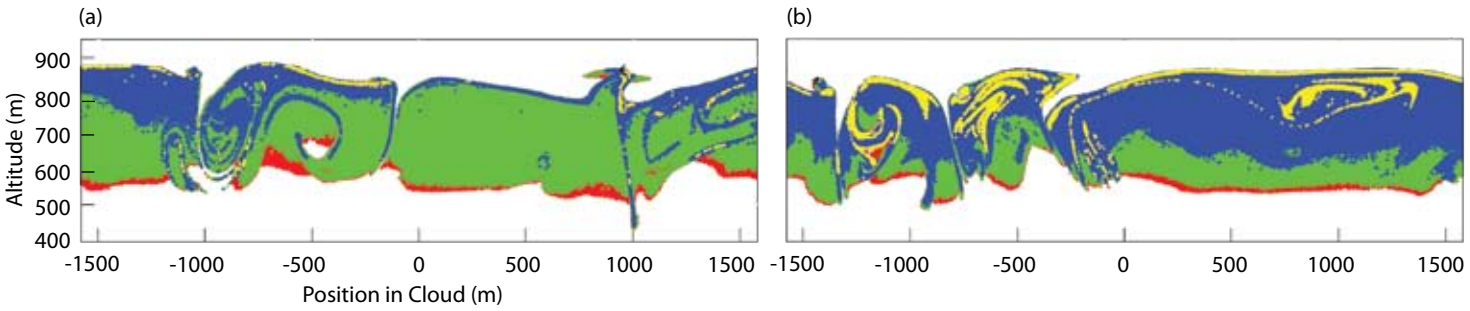
But the MTI didn't just hand over ready-made answers. It provided measurements of radiance (light intensity) that Chylek and his team had to translate into accurate information about aerosol optical depth. After extensive calibrations with ground observations, Chylek hit upon a method that turned out to be highly accurate. For more global atmospheric data, Chylek turned to NASA's satellite-mounted MODIS instrument, which images the entire Earth every 1 to 2 days. While NASA had its own way to calculate aerosol optical depth, Chylek's method reduced the error by a factor of 2 to 3. This led to surprising observations of aerosol behavior over the Indian Ocean, a long way from the Arctic but involving the same culprit: black carbon.

During the winter, pollution increases dramatically over the Indian subcontinent, affecting cloud formation. As expected, Chylek found that more aerosols produced more and smaller cloud droplets, reflecting more sunlight back into space. But at altitudes where icy cirrus clouds form, the result was, surprisingly, the opposite—fewer but larger ice crystals letting more solar radiation through to the surface. This puzzling reaction was caused by the water-attracting properties of black carbon, and between the recent growth of industry and the longstanding practice of burning wood and coal as household fuels, India produces a lot of soot. In the dance between aerosols and water vapor, water snubs the other aerosols, and clings to the soot, which has the additional power to initiate freezing at just a few degrees below zero. In contrast, water condensed around sulfate particles can remain liquid up to –40°C. So water freezes quickly onto soot, depletes atmospheric moisture, and leaves many aerosols without a condensation partner. Climate models were not including this reaction.

Chylek's and Dubey's work complement each other well. Dubey measures aerosols over a city here and a region there while flying at different altitudes, which leads to highly detailed results, but it would require years to combine those details into a global picture. Chylek's satellite observations lack the detail, seeing



Satellite observations near India were as expected for lower-altitude water clouds but surprising for higher-altitude ice clouds. Increased pollution during the winter created many smaller cloud droplets (purple) in water clouds. But human-generated soot led to fewer but larger droplets in ice clouds, allowing more solar radiation through.



only optical averages of a whole column of aerosols from surface to satellite, but allow analysis of the entire planet in just days. Together, their work produces the stuff that better climate models are made of.

Cloud Modeling

Jon Reisner sits in a bare, modest office inside a small, weather-beaten, prefab building, but he's connected to one of the most-sophisticated supercomputing architectures on the planet. As he explains the complexities of cloud modeling, he keeps an eye on line after line of code from a lightning simulation scrolling down one of his oversized computer screens.

Reisner, of Los Alamos' Earth and Environmental Sciences Division, has made recent breakthroughs in accurately modeling aerosol-cloud processes. His success lies in his willingness to use an approach assumed unsuitable by most of the atmospheric modeling community. It's called the Lagrangian method. As Reisner worked through the physics and math, he found that the common objections to the method were unfounded.

Modeling aerosols and clouds is chiefly about simulating particle behavior, something Los Alamos has been perfecting since its founding. From the protons and neutrons of nuclear reactions to the toxic chemical and biological plumes of potential terrorist attacks, if it involves particles, Los Alamos has probably modeled it. Of the two common computational approaches to particle modeling, the Lagrangian and the Eulerian, the latter has been favored by atmospheric modelers, and that fact has led to some problems in simulating aerosol-cloud interactions.

The mathematical differences between the Lagrangian and Eulerian methods are not easily translated into words, but to get a simplified idea, think of a two-dimensional grid, like a checkerboard, with checkers representing aerosol particles. Modelers break large problems into grids to make complex calculations of the whole problem easier. In a Eulerian simulation, if a checker moves out of its grid square to cross the corner where four squares meet, it is no longer considered a single checker but a fraction of a checker in each of the four grid squares it partially covers. The checker—aerosol particle—gets diffused, which results in problems within computer simulations, especially at a cloud's edge, which is a border between droplets and no droplets. This Eulerian edge problem can cause clouds to instantly vanish from a simulation when, in reality, they would have survived a day or two.

In contrast, the Lagrangian approach solves the edge problem by tracking the checker/particle across grid lines, always representing it as a single undiffused checker. This results in more-accurate tracking and location of particles and also enables better calculations of particle collisions, which are important because colliding cloud droplets merge to form large, falling raindrops—a process that is not yet well understood.

Reisner's method was so accurate that Dubey turned to him to model aircraft observations of the effects of soot pollution on clouds. After 20 other models had failed to simulate the data, Reisner's method successfully re-created the observed response of clouds to soot.

An Emerging National Security Mission

A precise understanding of aerosols will take many more years of scientific work, but the good news is that aerosols are very short-lived compared with greenhouse gases. If we stopped pumping CO_2 into the atmosphere right now, most of what's already there would linger for another 50 to 200 years and some for thousands. But aerosols stay aloft from a few minutes to 10 days. Once we better understand their climatic effects and decide on controls, we can deal with aerosols quickly—if everyone cooperates.

Earth's climate is an international problem, and international solutions require treaties. Treaties work only if we can verify compliance. Detecting and tracking man-made aerosols and greenhouse gases against the background noise of natural emissions is no easy task, but it's the same kind of task that Dubey, Chylek, Reisner, and others are undertaking to measure and model aerosols. The knowledge honed through their research will produce the expertise and tools needed to verify compliance with environmental treaties. Such treaties, like the Nuclear Test Ban Treaty of the past, may be integral parts of the future national security landscape.❖

—Anthony Mancino

Above: Cloud simulations made using a Lagrangian modeling approach. Red indicates the smallest droplets, followed by green, blue, and the largest, yellow. High aerosol concentrations (left) lead to many small droplets while lower concentrations (right) produce fewer larger droplets.