The greenhouse effect is a geophysical fact of life. Atmospheric gases such as carbon dioxide and methane trap and hold heat, enabling the earth’s biota to survive. Such gases warm the surface of this planet by about 33 degrees Celsius, from below freezing to a current average of about 17 degrees C. Models and analyses of global warming generally agree that most of the long-lived gases that human economic activity adds to the atmosphere make the earth warmer than it would otherwise be. Yet discrepancies between theory and observation persist. The predicted warming based on recent increases in concentrations of greenhouse gases is slightly more than the observed warming of the atmosphere. In addition, the warming trend in North America does not appear to follow the global pattern. What might account for these and other deviations of fact from theory?

The answer is ironic. In all probability, aerosols primarily composed of sulfates, themselves the result of commercial activity, enhance the ability of the atmosphere to reflect sunlight back into space before it can reach the planet’s surface and participate in the warming process. The sulfate particles, about 0.1 to one micron in diameter, are particularly concentrated over the industrial areas of the Northern Hemisphere. Their roles as contributors to acid rain, as irritants and as obscurers of such splendid vistas as the Grand Canyon have been known for years. But their capacity to cool by scattering sunlight has become a recognized force in climatic change only recently. Clearly, both the cooling effects of aerosols and the warming caused by greenhouse gases must be taken into account if we are to attain accurate climate models and effective industrial policies.

In theory, industrial aerosols are not the only particles that can contribute to cooling. Several kinds of aerosols exist.
4. Residing mostly in the lower troposphere, sulfate aerosol directly cools the earth by reflecting sunlight. The particles may also act as seeds for cloud condensation and increase the reflectivity, or albedo, of clouds.

5. When injected into the stratosphere by large volcanic eruptions, the sulfate compounds can help destroy the ozone layer.

6. The sulfate particles acidify rainwater, damaging lakes and plant life.
naturally. They do not, however, seem to be major factors causing change. Natural aerosols—mostly continental dust, sea salt and marine sulfate compounds—have probably remained roughly constant in their concentration, distribution and properties for at least a century. Thus, they would not have contributed to any observable alterations in climate. Volcanic aerosols have probably not added to long-term effects. The cooling trends precipitated by the gigantic eruptions of Tambora in 1815, Krakatoa in 1883 and Pinatubo in 1991 lasted only a few years.

In stark contrast, man-made aerosol compounds in the atmosphere have increased dramatically, primarily during the course of industrialization and most rapidly since about 1950. Of all the particulate pollutants humans create, climatologists have thus far focused much of their attention on sulfate compounds. That is because a large body of data, gathered in studies of acid rain, makes sulfates the best-understood aerosol. Other aerosol substances—soot from oil combustion, soil dust from desertification and smoke from slash-and-burn agriculture—may have an impact approaching the magnitude of that caused by industrial sulfur. Limited research findings, however, render the uncertainties in calculating those effects much greater.

As one might expect for complex systems such as the climate, determining the amount of cooling by sulfate aerosol is not a straightforward task. Many variables complicate the effort—among them, the amount of sulfur in the atmosphere, its distribution over the globe, the mechanism of aerosol formation, the degree of reflectivity of the particles and their effect on clouds. An accurate prediction also depends on making correct assumptions. Some early studies exploring the role of aerosols on climate did not do so. For example, one common and unsubstantiated supposition was that most of the haze outside cities was a "natural background" aerosol.

Another early, implicit assumption was that processes at the earth’s surface make most aerosol particles. But that conclusion is valid for only two kinds of aerosols: those introduced into

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the atmosphere by wind (such as sea salt and soil dust) and those arising directly from combustion (for example, industrial smoke or smoke particles from forest and grass fires). Studies during the past decade indicate that most sulfate aerosol originates from chemical reactions of sulfur gases discharged into the air. These reactions take place in the troposphere, that part of the atmosphere extending from the surface of the earth to an altitude of about 10 kilometers.

To calculate the increases of sulfur in the troposphere, climatologists rely on industrial emission rates. These rates act as excellent guides for estimating changes in the average concentration of atmospheric sulfate aerosol over time. Sulfur gases and the sulfate they make last only a few days in the troposphere, so that the average concentration of the substances is directly proportional to the product of the emission rate and the lifetime of the substances. Consequently, the primary effects must mirror the geographic distribution of the sources of sulfur.

More than two thirds of the troposphere’s supply of sulfur gases, mostly emitted in the form of sulfur dioxide (SO₂), is man-made. About 90 percent of that amount arises in the Northern Hemisphere. There human activity injects about five times the amount of sulfur gases emitted naturally. In the Southern Hemisphere, man-made emissions currently equal only about one third of natural emissions. The main natural carrier of reactive sulfur is dimethyl sulfide ((CH₃)₂S), or DMS, which originates from marine phytoplankton. In the absence of anthropogenic sources, DMS is thought to be the dominant source of submicron particles. A small amount of sulfur (as hydrogen sulfide or sulfur dioxide, or both) comes from volcanoes and from swamps and bogs.

The sulfur dioxide generally remains in the hemisphere in which it was produced. The thermal and chemical mixing of the two halves of the earth’s atmosphere requires about a year—far longer than the average lifetime of sulfur dioxide or the sulfate aerosol it produces. Although the hemispheres are essentially decoupled regions as far as aerosol distributions are concerned, the aerosols in the Northern Hemisphere may nonetheless influence the climate worldwide, just as regional cloud cover controls the average albedo, or reflectivity, of the earth.

About half the amount of sulfur gases is lost directly from the atmosphere; it is either washed out by rain or reacts chemically with plants, soil or seawater. The remainder goes on to oxidize with compounds in the troposphere and hence to produce aerosol particles. Indeed, almost all types of sulfur-containing gases are chemically reactive in the presence of oxidizing agents. The most important such agent is the hydroxyl (OH) radical.

The reactions that create sulfate aerosol can loosely be divided into clear-sky and in-cloud processes. In clear-sky processes, sulfur dioxide and DMS in the presence of water vapor react via a complex series of steps to produce gaseous sulfuric acid (H₂SO₄). The compound forms parti-
Sulfate aerosol is also produced in clouds. This pathway begins when sulfur dioxide dissolves into existing cloud droplets. There it may be oxidized by the small concentrations of aqueous hydrogen peroxide (H$_2$O$_2$) that form when two hydroxyl molecules combine. The oxidation reaction then forms sulfuric acid and its ammonium salts in solution. In the droplet the acid sulfate exists as a strongly hydrated form, in which water molecules are bonded to the sulfate. Evaporation removes some of the moisture. Because the sulfate clinging to water, the product of evaporation is a highly concentrated sulfate solution. The result is a submicron aerosol droplet that is chemically indistinguishable from the aerosol produced by gas-to-particle conversion.

The strong chemical affinity that sulfuric acid and its ammonium salts have for water is highly significant in terms of the aerosol’s ability to scatter light. When the tiny solution droplets mix with humid air (such as over moist land or oceans), they absorb moisture and grow. Larger particles scatter more visible light, thus explaining the increase in haze when humidity is high. At a relative humidity of 80 percent (the global average value for air near the ground), a given amount of sulfate produces about twice as much apparent haziness as it does during a low-humidity day.

Once formed through chemical reactions, sulfate particles in the troposphere can cool the climate in two ways: either directly, under clear skies, by reflecting away some incoming solar radiation, or indirectly, by increasing the reflectivity of clouds.

In the direct, or clear-sky, effect the sulfate aerosol particles scatter sunlight out of the atmosphere and into space; as a result, less solar radiation reaches the ground. There are two ways to estimate the fraction of incoming energy lost to space. One technique is to conduct detailed optical calculations based on particle sizes and refractive indices. An alternative and currently more reliable approximation is simply to make use of the observed correlation between the amount of aerosol in the atmosphere and the energy loss caused by scattering [see box on page 57]. These analyses indicate that at today’s levels man-made sulfate scatters about 3 percent of the direct solar beam. About 15 to 20 percent of this amount goes back into space, for a total loss of about 0.5 percent. The average reduction of sunlight, however, is actually about half this amount, because clouds cover about half the earth at any given time. On the ground, the deprivation of sunlight is calculated to be roughly about 0.2 to 0.3 percent.

Is this loss at all significant? The solar radiation reaching the layer of sulfate haze near the ground amounts to roughly 200 watts per square meter, so the implied loss amounts to about 0.4 to 0.6 watt per square meter. Because the Northern Hemisphere contains more aerosols, the average forcing there must be greater, probably around one watt per square meter. (Climatologists use the term “forcing” to refer to the effect of factors external to the atmosphere and oceans on the changes in the planetary energy balance.) Such a loss of incoming energy may seem small, but it is not inconsequential. The present-day increases of carbon dioxide resulting from human activity amount to a gain of 1.5 watts per square meter in the planet’s heat balance. (When other greenhouse gases such as methane and nitrous oxide are considered, the increase is about two to 2.5 watts per square meter.) Hence, the cooling caused by sulfate aerosol is...
comparable in magnitude to the heating caused by carbon dioxide, at least in the patches of haze concentrated over industrial regions.

Needless to say, these calculations are crude. To quantify more precisely the aerosol effect and to describe its geographic distribution, researchers at Stockholm University and the University of Washington used a meteorologic model developed at the Max Planck Institute for Chemistry in Mainz. The description, which details the chemical production and wind-driven transport of particles generated from anthropogenic sulfur dioxide, enabled the workers to produce a map of the change in heat balance caused only by the direct effect of anthropogenic sulfate. This model showed three large masses of haze in the Northern Hemisphere. One mass, over the eastern U.S., creates losses of solar radiation of more than two watts per square meter. The two others, over Europe and the Middle East, reflect up to four watts per square meter. The average over the Northern Hemisphere, based on sulfur dioxide emissions recorded in 1980, is 1.1 watts per square meter, happily close to the crude calculation above.

The second, indirect way sulfate aerosol cools the earth is by influencing the albedo of clouds. When in clouds, some of the sulfate particles act as nuclei for condensation. The density of cloud-condensation nuclei determines the number and the size of cloud droplets. For a given amount of condensed water, the number density in turn affects the albedo of the cloud. A 30 percent rise in cloud albedo only over the world’s oceans would be sufficient to counteract the average warming by anthropogenic carbon dioxide increases during this century.

Unfortunately, this indirect effect of the sulfate particles has thus far resisted reliable quantification. Although observations show that cloud-condensation nuclei are greatly enhanced over industrial regions, investigators do not know how differences in the number of nuclei relate to the changes in the amount or mass of anthropogenic aerosols. As a result, estimating the magnitude of the indirect aerosol forcing is not yet possible. Satellite observations suggest that the effect is not huge, although theoretical analyses permit it to be comparable to direct forcing.

In view of the fact that the ability to model completely the meteorologic effects of aerosols is limited, one may wonder whether aerosol cooling is real. In particular, one may ask whether the aerosol cooling is evident in the observational record. The most straightforward way to answer this question is to compare the changes in the Northern Hemisphere with those in the Southern Hemisphere. As a whole, the globe has warmed by about 0.5 degree C during the past 100 years [see “Global Warming Trends,” by Philip D. Jones and Tom M. L. Wigley; SCIENTIFIC AMERICAN, August 1990]. If the enhanced greenhouse effect (that is, the additional warming caused by human activity) is the sole mechanism for climatic forcing, then the Northern Hemisphere should warm a bit more quickly than the Southern Hemisphere. The Southern Hemisphere holds most of the world’s oceans and hence has more inertia with respect to thermal changes.

Yet the observations show otherwise: since 1940 the Northern Hemisphere has warmed more slowly. In fact, the strong warming trend that occurred earlier this century in the Northern Hemisphere ceased around 1940 and was not renewed until the mid-1970s, even though industrial emissions of greenhouse gases continued to rise over the entire period. This reprieve in warming may have resulted from the countacting properties of sulfate aerosol, at least to some extent. Although the changes broadly parallel the hypothesized aerosol cooling, they are not enough to prove a causal relation. (Indeed, the lack of a marked difference between the warming trends in the two hemispheres throughout the 20th century imposes an upper bound on the total magnitude of the aerosol forcing, which implies that the cloud albedo contribution has been small.)

Another piece of circumstantial evidence comes from an analysis conducted by the United Nations Intergovernmental Panel on Climate Change (IPCC). In 1990 the panel pointed out a discrepancy between the observed global mean temperature changes and the predictions made by climate models. The models suggested that the world should have warmed somewhat faster than the record indicates. Sulfate aerosol may help explain the discrepancy.

To see why, we need to introduce the concept of “climate sensitivity.” In computer simulations of the climate, investigators double the atmospheric carbon dioxide concentration and then allow the climate system to adjust to the new (warmer) steady state. The change in global mean temperature is a measure of the sensitivity of the global average temperature to external forcing. The IPCC has given a “best guess” value of 2.5 degrees C for this quantity, although the sensitivity may in fact range from 1.5 to 4.5 degrees C. When observations are compared with the results from climate models designed to estimate specifically the time-dependent response to observed changes in greenhouse-gas forcing, the implied climate sensitivity is found to be a little less than 1.5 degrees C. In other words, the empirical estimate of the climate sensitivity gives a value more than a full degree below

SULFATE AEROSOL sampled from the atmosphere was photographed through an electron microscope. The particles are about 0.1 micron in diameter.
the IPCC’s best guess and slightly below the expected range.

These numbers suggest that the current global warming induced by greenhouse gases may have exceeded the observed 0.5 degree C rise and been offset by some kind of cooling process. Natural variability of the climate could account for the cooling. Alternatively, external factors may be responsible. The aerosol effect is an obvious candidate. Indeed, factoring aerosol cooling into models yields a value for climate sensitivity that is a little above the IPCC’s best guess but well within the expected range. Unfortunately, none of the conclusions is sufficiently convincing to allow us to jump out of the bath-tub crying, “Eureka!”

Although subject to considerable quantitative uncertainty, the evidence clearly indicates that aerosols have a significant influence on the climate, comparable to that produced by greenhouse gases. In fact, from 1880 to 1970, aerosol cooling may be more or less have canceled out the enhanced greenhouse effect in the Northern Hemisphere. (Since 1970 emissions of greenhouse gases have increased more rapidly than have those of aerosol particles.) The cooling caused by aerosols may even dominate in some areas. Recent work by Jeffrey T. Kiehl and Bruce P. Briegleb of the National Center for Atmospheric Research in Boulder, Colo., suggests that aerosols produce a net cooling in local regions of the eastern U.S., south central Europe and eastern China.

A crucial complication, however, is hidden in the use of the words “cancel out.” The term is deceptive. Aerosol cooling and the greenhouse effect have characteristics that prevent them from neatly offsetting each other. First, the cooling and warming occur mostly over different parts of the world. As we mentioned, sulfate cooling happens primarily over industrial zones in the Northern Hemisphere. Although carbon dioxide spreads throughout the atmosphere, greenhouse forcing is more potent over the subtropical oceans and deserts.

Both types of forcing also differ temporally. The heat-trapping property of carbon dioxide varies only moderately during the course of a day and throughout the year. In contrast, the aerosol effect has a distinctive diurnal and seasonal character. It acts more vigorously in the summer and, of course, operates only during daylight hours. Thomas R. Karl of the National Climatic Data Center and his co-workers have shown that the U.S., the former Soviet Union and China all have displayed increases in annual average minimum temperatures but no increases in the maximum temperatures. It is possible, therefore, that aerosols may now be canceling out greenhouse warming during the day (when temperatures are highest) but not at night (when temperatures are usually lowest).

How should one regard the evidence garnered to date for aerosol cooling? A good way to judge is to compare it with the enhanced greenhouse effect. Although the IPCC recommends large-scale cuts in emissions of carbon dioxide, it has not been able to say conclusively that changes in greenhouse-gas concentrations have caused the observed global warming. A definitive verdict is difficult because the magnitude of the effect to date is roughly comparable to the natural variability of the climate. In other words, the signal is about the same strength as the background noise.

Precisely the same situation applies to aerosol forcing of the climate. Climatologists have not yet found the “smoking gun” that would prove beyond doubt the existence of sulfate cooling. Yet the strong theoretical basis of the aerosol effect, the consistency of the data with expectations and the lack of any counterevidence give us considerable confidence in its reality. Still, two large areas of uncertainty limit our predictive capability: understanding the fundamental physics of global climatic change and forecasting the levels of future emissions of sulfur dioxide. Right now the estimated uncertainty in forcing caused by the best-understood anthropogenic aerosol—sulfates—is much greater than the uncertainty in forcing created by greenhouse gases. For sulfates, the amount of cooling ranges by a factor of two; for greenhouse gases, the degree of warming is known to within roughly one tenth to one fifth.

Nevertheless, we can make a few general predictions. Because anthropogenic sulfate aerosol is confined for the most part to specific parts of the Northern Hemisphere, greenhouse warming should proceed relatively unabated in the Southern Hemisphere (and in more rural parts of the Northern Hemisphere). The IPCC forecast of a sea-level rise of a few tens of centimeters over the next 50 years therefore remains reasonable. A substantial fraction of this rise is associated with the global-scale thermal expansion of the warming water. Other repercussions are somewhat harder to predict, because they depend on the regional details of the combined aerosol and greenhouse forcing.

Reducing the emissions of carbon dioxide and sulfur dioxide would have two contrasting outcomes. Because the
How Much Light Do Aerosols Reflect Away?

Atmospheric sulfate aerosol scatters light in all directions. About 15 to 20 percent of the light is scattered back into space. The backscattering constitutes the direct effect of atmospheric aerosol on incoming radiation. The light-scattering efficiency of aerosol, represented by the Greek letter alpha ($\alpha$), is high, even at low humidity: each gram represents an area of about five square meters. Moisture increases the scattering by making the aerosol expand. At the global average relative humidity, the efficiency doubles, to almost 10 square meters per gram. One can use this value to estimate the magnitude of the direct effect of anthropogenic sulfate.

The rate at which light is lost from the solar beam is defined by the scattering coefficient, represented by the Greek letter sigma ($\sigma$), expressed in units of per meter. This value is determined by the amount of aerosol mass, $M$ (in grams per cubic meter), multiplied by the light-scattering efficiency: $\sigma = \alpha M$. When both sides of this equation are integrated over altitude, $z$, a dimensionless quantity called the aerosol optical depth and represented by the Greek letter delta ($\delta$), results:

$$\int_0^\infty \sigma \, dz = \delta = \alpha \int_0^\infty M \, dz = \alpha B$$

Here $B$ is the world average burden of anthropogenic sulfate aerosol in a column of air, in grams per cubic meter. The optical depth is then used in the Beer Law (which describes the transmission of light through the entire vertical column of the atmosphere). The law yields $I/I_0 = e^{-\delta}$, where $I$ is the intensity of transmitted radiation, $I_0$ is the incident intensity outside the atmosphere and $e$ is the base of natural logarithms. In the simplest case, where the optical depth is much less than 1, $\delta$ is the fraction of light lost from the solar beam because of scattering. The question, then, is just how large $\delta$ is or, more properly, that part of it that results from man-made sulfate.

This global average burden of anthropogenic sulfate aerosol can be estimated by considering the entire atmospheric volume as a box. Because the lifetime of sulfate aerosol is short, the sum of all sulfate sources, $Q$, and its lifetime in the box, $t$, along with the area of the earth, determine $B$:

$$B = \frac{Q t}{\text{area of the earth}}$$

About half the man-made emissions of sulfur dioxide become sulfate aerosol. That implies that currently 35 teragrams ($3.5 \times 10^{12}$ grams) per year of sulfur in sulfur dioxide is converted chemically to sulfate. Because the molecular weight of sulfate is three times that of the elemental sulfur, $Q$ is about $(3)(3.5 \times 10^{12})$ or $1.1 \times 10^{14}$ grams per year. Studies of sulfate in acid rain have shown that sulfates persist in the air for about five days, or 0.014 year. The area of the earth is $5.1 \times 10^{14}$ square meters. Substituting these values into the equation for $B$ yields about $2.8 \times 10^{-3}$ gram per square meter for the burden.

This apparently meager amount of material produces a small but significant value for the aerosol optical depth. Using the value of scattering efficiency ($\alpha$) of five square meters per gram and a factor of two for the increase in scattering coefficient because of relative humidity, the estimated anthropogenic optical depth becomes $\delta = 5 \times 2 \times (2.8 \times 10^{-3}) = 0.028$. This value means that about 3 percent of the direct solar beam fails to reach the earth's surface because of man-made sulfate. A smaller amount—perhaps 0.15 percent, or about 0.5 percent—is thus lost to space. This scattering operates over the noncloudy parts of the earth. About half the earth is cloudy at any given time, so that globally 0.2 to 0.3 percent is lost.

FURTHER READING


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