A skeptical evaluation of the sulfate problem

James W. Sawyer

Sulfur dioxide was written into law as the cause of dirty air, but all that has become clearer is that SO$_2$ is not the only nor even the major culprit. Regardless of laws, we have only begun to solve the complex riddle called air pollution.

This paper approaches the so-called sulfate issue in a skeptical spirit. By this approach I hope to at least present a disinterested summary statement of the sulfate issue: (i) its origins, (ii) the significance of anthropogenic sources, (iii) the toxicological and epidemiological evidence, (iv) the associated acid rainfall problem and its effects, (v) the problem of visibility reduction, (vi) the practicability of stack gas scrubbing, (vii) the problems of costs and benefits, (viii) the past and future contributions of mathematical modeling, and finally (ix) the problem of making reasonable policy dealing with the sulfate issue.

ORIGINS

The history of the sulfate problem is perhaps stranger than that of any other pollutant. The Air Quality Act of 1967 required the Secretary of HEW to develop air quality criteria which would reflect the latest scientific knowledge useful in indicating the kind and extent of all identifiable effects on health and welfare which may be expected from the presence of an air pollution agent. Welfare was to include impacts on aesthetics, plants, wildlife, materials, visibility, weather and climate, as well as impacts on economic values and personal comfort.

HEW went to work on this mandate immediately. The criteria document summarizes data on sulfur dioxide, sulfur trioxide, and various sulfate and sulfur salts. In addition to sulfur oxides, the document also addresses the particular hazards of the sulfates and sulfuric acid mists. The problems of acid rain, materials corrosion, and effects on vegetation and animals are all discussed. The summary of the criteria document reiterates the importance of sulfates, sulfuric acid, and particulates as critical components of the effects of sulfur oxides pollution.

However, when the conclusions were spelled out relating effect to pollutant level, the results were expressed as equivalent ambient sulfur dioxide concentrations (and sometimes in terms of an associated particulate level). This method of reporting neglected the fact that the scientists involved were quite evidently discussing that complex of problems which we now refer to as the sulfate problem. It seems plausible to suggest that, just as we are confused about exactly what the problem is now, the scientists found it hard to understand then. Perhaps this is at least one explanation why they took the easy way out, and reported their results in terms of equivalent sulfur dioxide concentrations.

R. E. Trumbule, et al. of the Congressional Research Service, in reviewing the history of the sulfate problem, suggests three other possible explanations for reporting the hazards in terms of equivalent sulfur dioxide levels. First, it provided a seemingly uniform comparative basis for reporting the results. Second, nearly all anthropogenic emissions of sulfur oxides are in sulfur dioxide form: thus controls would be in terms of sulfur dioxide emissions. Third, at that time it was thought that the transformation of sulfur dioxide to sulfates and sulfuric acid occurred at fairly constant rates.

The first two explanations reduce the problem of administration and implementation enormously, and are understandable. The third, in a sense, rationalizes the other two. If the third were not true the first two explanations are essentially unsupportable.

We now know that the third is not true. Rather than the transformation rate being constant, the rate of oxidation of sulfur dioxide to various sulfates varies between 0.1 and 30 percent per hour, a factor of 300. Moreover, the conversion is catalyzed by photochemical smog and varies with sunlight intensity, ambient temperatures, humidity, and the presence of particulate matter.

It is evident enough from this short narrative of the events that a completely understandable bureaucratic error led policymakers in both state and federal government into taking the wrong fork in the road to environmental improvement. Power companies spent large sums of money developing a rationale for tall stacks, or intermittent control systems, as a method of reducing sulfur dioxide concentrations. Researchers spent large quantities of money developing elaborate mathematical models to predict sulfur dioxide concentrations. And EPA spent large quantities of money supporting the mathematical modelers.

Initially it was relatively easy for power companies to meet the sulfur dioxide emission limitations by switching to sources of low sulfur oil and gas. When the fuel crisis of 1973 occurred, however, they were hurt badly. With both oil and gas becoming increasingly scarce, they had to either switch to expensive low sulfur coal or else expensive (and at that time unreliable) stack gas scrubbing.

Enough progress was made in reducing ambient sulfur dioxide concentrations in many U.S. locations for scientists to discover that the relationship between local sulfate concentrations and local sulfur dioxide emissions was much more complex than originally anticipated. Whether local sulfur dioxide concentrations stayed high or fell off radically, local sulfate concentrations stayed high—alarmingly high as far as epidemiologists were concerned. Moreover, sulfates seemed to be ubiquitous. In the Northeast it was a simple task to find high ambient concentrations in remote rural locations far removed from any recognizably large sources of sulfur dioxide.

The understandable bureaucratic error, then, was costly to us all. After some six to seven years we are almost where we started as far as sulfur compound air quality is concerned. The costs to power companies—and the public—has been very large.

ANTHROPOGENIC AND NATURAL SOURCES OF AIR-BORNE SULFUR COMPOUNDS

Anthropogenic sources account for about 1/4 to 1/3 of the sulfur (in one chemical form or another) in the planetary atmosphere; estimates range from 100 to 150 million tons per year. Some 90 percent of these anthropogenic sources are located in the northern hemisphere. Thus about 1/2 to 1/3 of the airborne sulfur (compounds) in the northern hemisphere is from anthropogenic sources. The anthropogenic sources of airborne sulfur compounds are on land, which comprises only a fraction of the surface area of the northern hemisphere. And of course the anthropogenic sources tend to be concentrated in relatively small regions, for example the northeastern section of the U.S. It has been estimated that 80-90 percent or more of the sulfur oxides present in cities is anthropogenic.

As a reasonable first estimate, something on
the order of 80 percent of the sulfate compounds present in the northeastern U.S. are from anthropogenic sources. While this is a rough estimate, its plausibility is easily established. The non–urban sulfate concentrations over most of the U.S. west of the Mississippi are about 1-3 micrograms per cubic meter. This is a region where there are relatively few power plants, industrial plants generally, and low population densities. It seems reasonable to use, say, 2 micrograms per cubic meter as “natural” background sulfate concentrations. In the industrial northeast, where there are large population densities, much traffic, and large industries, ambient rural sulfate concentrations range from 7 to greater than 9 micrograms per cubic meter. These combined data suggests that about 57% to 79%, or 70% to 77%, of the rural ambient sulfate is from anthropogenic sources. A figure of 75-80 percent as an approximate anthropogenic source sulfur compound contribution for the northeast seems a good rule of thumb.9

The next significant point is to establish those anthropogenic sources which are of greatest significance. The combustion of coal and petroleum products accounted for about 80 percent of total sulfur dioxide emissions in 1966.10 (This figure is aggregated over the U.S.). Sulfur oxide emissions have been growing at about 4% annually, and at an apparently far higher rate during the sixties for public utilities than other industries.11 The bulk of airborne sulfur compounds arise from power plants.12 There is a slight caveat here. In central cities and near heavily traveled highways, near-ground level air tends to contain most of its sulfate in the form of sulfuric acid mist, the product of automobiles and their catalytic removal systems. This effect is, however, highly local.

Later on in the paper I shall briefly refer to NOx emissions and their associated problems. The incremental increase in ambient NOx also appears, on cursory examination, to be intimately linked to increasing electric power production.13 More than 55 percent of anthropogenic NOx production in the U.S. originates from stationary sources, and 93 percent of these emissions are from fossil fuel combustion.14

Direct Effects of Sulfates on Health: Toxicological and Epidemiological Evidence

Dr. Carl Shy, in testimony before the House Subcommittee on the Environment and the Atmosphere, has summarized the state of knowledge concerning the health effects of acid sulfates.15 A partial quote from Shy’s testimony is as follows:

"When gaseous sulfur dioxide is emitted into the atmosphere, this gas reacts with water to form sulfuric acid aerosol and with various metallic ions to form metallic sulfates. These acid-sulfate aerosols are in the fine particulate, respirable size range.

1. Experimental studies of pulmonary flow resistance in animals reveal that sulfuric acid, various metallic sulfates and mixtures of sulfur dioxide with particles in high humidity are more potent and more irritating to the airways than equivalent concentrations of sulfur dioxide alone.

2. Inhalation of sulfate aerosols increases pulmonary flow resistance in animals at concentrations at which inert aerosols have no effect.

3. In humans, experimental studies on healthy volunteers demonstrate a synergistic effect of sulfur dioxide and ozone. When volunteers breathed 0.37 ppm sulfur dioxide alone for 2 hours, no changes in lung function occurred. However, inhalation of 0.37 ppm sulfur dioxide mixed with 0.37 ppm ozone produced a 40 to 50 percent decrease in pulmonary flow rate. Ozone alone at this concentration had a much smaller effect—causing a 10 to 15 percent decrement in lung function."

Shy’s statement is persuasive in making the case that some thing or things about mixtures of sulfur dioxide, water, particulates, and oxidants, are harmful to the health of human beings. At the current time the causal agent is thought, though not thoroughly verified, to be sulfate compounds. Thus from a recent NAS report:16

... adverse health effects observed in humans are more likely attributable to transformation products of sulfur dioxide (sulfuric acid, sulfites, and sulfates, all indexed by particulate sulfate measurements) than to sulfur dioxide itself. These studies further suggest that adverse effects may be occurring even in communities that have not exceeded primary air quality standards for sulfur dioxide and total suspended particulates. Sulfur thresholds for adverse effects were estimated to be from 8 to 10 µg/m³ for 24-hour exposures. These estimates clearly require verification, so if they are correct, the sulfate health threshold is exceeded in a large portion of the urban areas in the entire eastern half of the United States for half the year or more. Nevertheless, in most eastern urban areas, primary air quality standard for sulfur dioxide—the precursor of manmade sulfates—has been achieved.

Epidemiology is the science that deals with the incidence, distribution, and control of a disease in a population. Thus epidemiological studies do not necessarily study the physiologic response of the human body to the toxin, but rather emphasize the incidence and distribution of some response, say morbidity, to something which might be identified as a toxin, say sulfur dioxide or sulfate.

The most famous—or infamous, depending on your point of view—epidemiologic study made on the complex of sulfate issues is the CHESS study (Community Health and Environmental Surveillance) study made by EPA.17 The CHESS study gets the credit for suggesting that the sulfur dioxide environmental problem might well be something far more complex, namely that which is now referred to as the sulfate problem.

The CHESS study suggests that the effects of sulfates on human health are the following: (1) at daily average sulfate concentrations of 0-10 micrograms per cubic meter asthma attacks increase in frequency; (2) at daily average concentrations exceeding 25 micrograms per cubic meter the elderly experience aggravation of heart and lung disease; (3) at current average concentrations exceeding 25 micrograms per cubic meter daily mortality increases; and (4) when sulfate concentrations exceed approximately 13 micrograms per cubic meter for several years, acute lower respiratory disease increases in children.18 If the reported health effects are correct, sulfate concentrations are already sufficiently high in the northeast section of the country that considerable chronic health problems are probably already occurring.19

Most physiologists and epidemiologists agree that while the presence of sulfur dioxide does produce a clear physiologic response in humans, sulfur dioxide as a compound is probably relatively innocuous at ambient concentrations in the U.S.

It is important to emphasize the intellectual difficulty here. It is clear that the presence of sulfur oxides along with other pollutants can be associated with