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ACID RAIN: THE IMPACT OF LOCAL SOURCES

By

Paul Spaite, M. P. Esposito, M. F. Szabo, and T. W. Devitt

November 24, 1980

Prepared for

UNITED STATES DEPARTMENT OF ENERGY
Morgantown Energy Technology Center
Morgantown, West Virginia

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SUMMARY

Little is known about the precise origin of acid rain precursor pollutants, especially with respect to the relative importance of local versus distant sources. Many have assumed that acid rain is predominantly a problem of long-range transport of pollutants from large fossil fuel combustion sources, namely coal-fired utilities. However, close examination of fuel use information and source emission characteristics in the Adirondacks, Florida, and California suggests that local oil burning and automotive sources may be major contributors to the occurrence of acid rain in these areas. This report describes the possible role of local combustion sources in the production of acid rain, discusses the implications of the findings, and their relevance to alternative control strategies for acid rain.

Oil-fired boilers, especially the smaller commercial, industrial, and residential units, produce at least 3 to 10 times as much primary sulfate per unit of sulfur content as coal-fired units. Moreover, oil-fired units emit comparatively large quantities of catalytic compounds capable of rapidly converting still more sulfur oxide to sulfate in the atmosphere. Thus, in areas where large quantities of oil are burned, the direct impact from locally generated sulfates may equal or even exceed that produced by "imported" sulfates derived from distant coal-burning sources.

Fuel consumption data show that large quantities of oil are being consumed in areas experiencing acid rain. Forty percent of the residual and 36 percent of the distillate oil burned in the United States is consumed in the eight-state area surrounding the Adirondacks. California is the next largest oil-consuming area and Florida is third.

Nitric acid is responsible for about 30 percent of rainfall acidity in the Northeast and Florida, and for about 30 to 75 percent of the rainfall acidity in California. Nitrates are believed to be formed from nitrogen oxides emitted principally by mobile and stationary combustion sources. In California, these NO_x emissions clearly arise from local sources; in Florida and the Northeast, they may be generated by both local and remote sources.

Application of flue gas desulfurization to the 50 largest coal-fired SO_2 emitters east of the Mississippi would cost about $12.5 billion and may not achieve the desired result if it is confirmed that local sources are significant contributors to acid rain. Local source control strategies such as ammonia injection, desulfurization, metal catalyst removal, and strict operating requirements may be needed to reduce $SO_2/SO_4^=$ emissions from oil-fired boilers. Advanced NO_x control techniques and conservation for both stationary and mobile sources may also be necessary. Residual oil backout of power plants would receive more serious consideration.

1. INTRODUCTION

Within the past decade increased attention and resources have been dedicated to understanding the origin, causes, and effects of air pollution. Acid rain is a relatively recent term that has emerged from this effort. It was coined to better describe how gaseous pollutants discharged into the air return to impact man's terrestrial environment. With improved measurement systems we are beginning to better understand the extent of the phenomenon, its severity, and the change in areas of impact. The occurrence of sulfates and nitrates in all acid rain has lead to the apparently obvious conclusion that sulfur oxide and nitrogen oxide air pollutants are the precursors of acid rain. Very little is known, however, about the precise origin of these pollutants or even the relative importance of local versus distant sources. In the absence of good data and information to guide our understanding of the acid rain phenomenon, we may have been too quick to assume that acid rain is predominantly a problem of long-range transport of pollutants. In fact, more recent studies suggest that acid rain may be a predominantly local phenomenon. This paper summarizes an ongoing study which attempts to look at all of the available data and information that could help establish the cause and sources of acid rain and lead to effective control strategies.

2. OCCURRENCE OF ACID RAIN IN THE UNITED STATES

The term "acid rain" denotes wet precipitation with a pH value below the normal pH of distilled water saturated with carbon dioxide (pH 5.6 to 5.7). This is also the pH level that many assume to be associated with unpolluted rain or snow. The term "acid rain" now also has a broader denotation covering all forms of precipitation with low pH that is perceived as contributing to acidification of surface waters, affecting fish populations and causing other environmental impacts.

The acidity of precipitation and the acidification of surface waters are attributed to increased emissions of sulfur and nitrogen oxides from anthropogenic sources. These emissions arise primarily from the combustion of fossil fuels (coal, oil, natural gas, gasoline), although other activities such as metal smelting, oil refining, and phosphate mining may also be important.

It is widely accepted that combustion of fuel is a dominant contributor where acid rain has been observed on a broad scale. The potential of fuel combustion, as opposed to other sources, for widespread impact is indicated in Table 1, which presents estimates of annual emissions.

TABLE 1. NATIONWIDE U.S. EMISSION ESTIMATES, 1977, 10^6 METRIC TONS[1]

Source Category	SO_x	NO_x
Transportation	0.8	9.2
Stationary Fuel Combustion	22.4	13.0
Industrial Processes	4.2	0.7
Other	0	0.2
Total	27.4	23.1

The mechanisms whereby the emitted pollutants eventually contribute to formation of acid rain are not well understood. It is generally believed that the oxides of sulfur and nitrogen are emitted to the atmosphere in completely oxidized form as primary sulfates or nitrates, or are emitted in partially oxidized form (SO_x, NO_x) and converted to their fully oxidized counterparts through atmospheric reactions involving other simple molecules such as water, ozone, and oxygen. The exact reaction sequence is uncertain. The theoretical rate of conversion of SO_2 to SO_4= is quite low, but conversion is believed to be accelerated in the atmosphere by catalyst pollutants such as vanadium, manganese, iron, copper, zinc, and possibly carbon[2-5]. Relatively high humidity (>75 percent) is also believed to be an important contributing factor in acid rain formation[2-5]. Seasonal variations in pH level, which are observed in the eastern states and in California, appear to be related to seasonal storm patterns. In the East, pH lows occur in the summer and are often associated with thunderstorms and showers arriving on cold fronts and high-pressure air masses[6,7]; in California, the pH lows often accompany winter storms[8].

Acid rain is reportedly occurring in many parts of the United States and in the eastern regions of Canada including Ontario, Quebec, and Nova Scotia. In the United States, acid rain is reported in New York, Pennsylvania, New Jersey, and New Hampshire. It is also reported in other parts of the country, including Florida, California, Washington, Colorado, Tennessee, Minnesota, and the Carolinas. In addition, low-pH rain has been reported in many cities. New York, Chicago, Philadelphia, Louisville, Evansville, Birmingham, Providence, and Boston have had precipitation with a pH less than 4. The mean pH of rain in Pasadena is 4.4. In Seattle it is 4.5 and in San Francisco it is 4.9. Acid rain also has been reported in Los Angeles.*

Although such reports might be construed as indicating that acid rain is so pervasive as to defy analysis of the causes, examination of information from three areas (the Adirondack Mountains, Florida, and California) where low pH rains have been observed suggests interesting clues as to possible relationships. These are discussed in the sections that follow.

*Reference 6 through 17 discuss occurrence of acid rain in the United States.

3. CAUSES OF ACID RAIN: LONG-RANGE VERSUS LOCAL SOURCES

LONG-RANGE TRANSPORT -- THE NORTHEAST SCENARIO

The northeast portion of the United States is believed to be the most strongly impacted by acid rain and acidification of surface waters. In the search for causes of this problem, it has become generally accepted that acidity in this much-studied region is primarily attributable to discharges from coal-burning power plants that emit large amounts of sulfur dioxide and small amounts of sulfate ion from tall stacks. It is believed that such discharges are transported over hundreds of miles and that during this transport, SO_2 and NO_x are transformed to sulfate and nitrate ions, which precipitate as acid rain at distant receptors. This logic was used to explain acid rain and acidification of lakes and streams in Scandanavia, where there are few local sources to contribute to an apparent trend to lower pH in precipitation and surface waters. This proposed explanation may be in part responsible for application of the same logic to explain acid rain in the northeastern United States and parts of Canada.

Various maps that have been generated to identify areas of low pH or high-atmospheric sulfate concentrations in northeastern United States generally indicate that highest impacts occur in eight states (Vermont, New Hampshire, Massachusetts, Connecticut, Rhode Island, New Jersey, Pennsylvania, and New York). These are shown in Figure 1 as the shaded area inside the isopleth for pH 4.52. The remote sources thought to be affecting this area are coal-fired power plants as far away as the Ohio River Valley. These plants are of 500-MW capacity or larger and use stacks 1,000 feet tall and taller to disperse the effluent gases over large areas. In the course of long-range transport, the SO_2 and NO_x emitted from these sources are thought to be slowly transformed into sulfates and nitrates, which are eventually deposited hundreds of miles away in lakes and streams to the Northeast, for example in the Adirondacks. A small amount of the primary sulfates formed in midwestern coal-fired boilers are deposited near the point of generation and make no significant contribution to acid rain in the Adirondacks.

LOCAL SOURCES -- ANOTHER POSSIBLE CAUSE

Numerous reports of the effects of local sources on acidification of the environment have appeared in recent literature. Local sources, including two thermal power generating stations and a petroleum refinery, are believed to be the major cause of acidification in Halifax County, Nova Scotia. Acid rain observed near Seattle, Washington has been linked to a nearby smelter[15]. Acid rain in Florida and California appears to result from local SO_x and NO_x emissions rather than from long-range transport.

Local oil-burning sources are especially suspect for two major reasons:

1. Burning of both distillate and residual oil produces large quantities of primary sulfates, which are formed in the furnace and need not undergo further chemical reaction to participate in formation of acid rain. Small residential and commercial boilers emit a much higher percentage of primary sulfates than do utility oil-fired boilers[18].

1965 - 1966

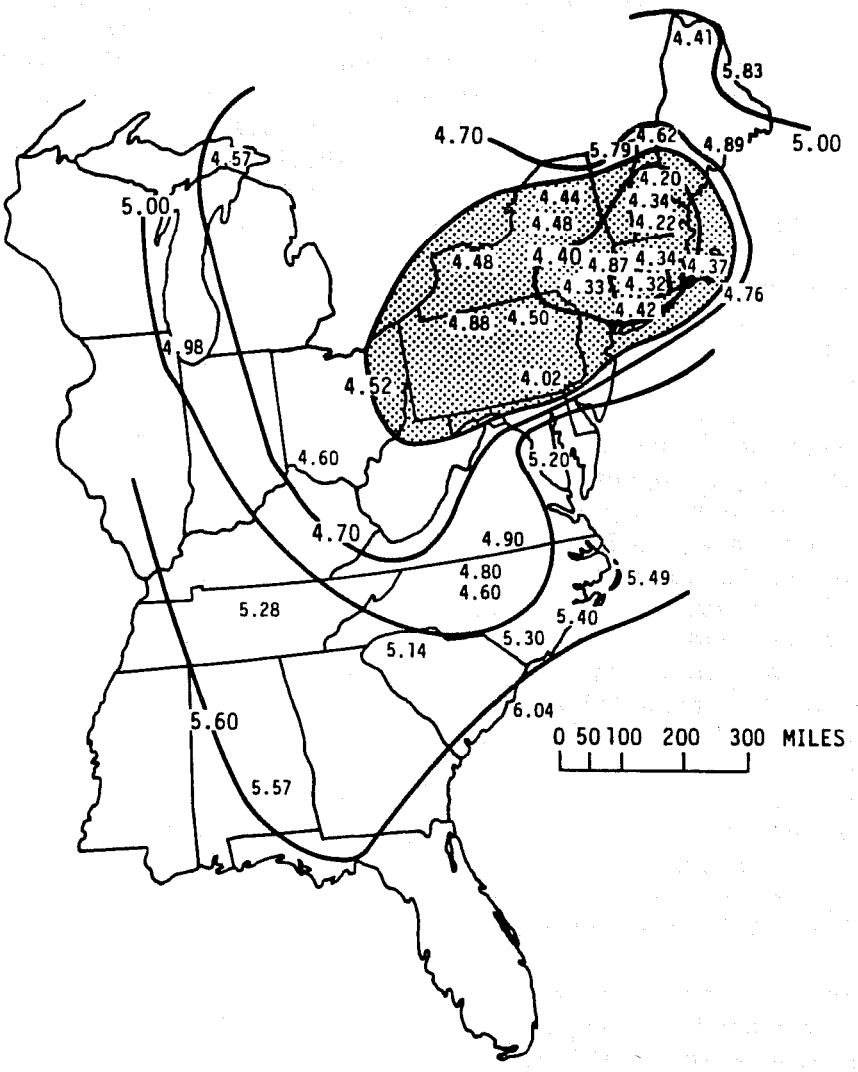


Figure 1. Acidity of precipitation in the eastern United States in 1965-1966[9].

2. Burning of residual oil releases large quantities of finely divided catalytic materials, such as vanadium and carbon, which can catalyze reactions in the plume and can further catalyze the transformation of sulfur dioxide to sulfate as these substances remain suspended in the atmosphere[2].

Where large quantities of oil are burned, the direct impacts of local oil burning may exceed those that would be produced by burning equivalent amounts of coal. In fact, the preliminary results of atmospheric modeling suggest that 50 percent or more of the sulfates impacting locally are produced locally.

Reports of studies implicating remote sources in acid rains in the Northeast usually refer to the possible contribution of local sources also. In the absence of any serious attempt to identify and quantify the local impacts, however, such comments have the effect of a minor qualification of the principal findings. Some of the evidence for local sources is considered in Section 4.

4. THE EVIDENCE FOR THE IMPORTANCE OF LOCAL SOURCES

In this investigation of the role of local sources in acidification, we have focused on the northeastern United States, California, and Florida. These areas present different settings for analysis of possible contributing causes. It is likely that the conventional theory implicating distant sources in the production of acid rain in the northeastern United States would not be applicable to California and Florida; data from the latter two states, therefore, offer promise for development of alternative theories.

THE NORTHEASTERN UNITED STATES

The sources contributing to acid rain in northeastern United States are depicted in Figure 2. Local oil-burning sources are shown along with remote coal-fired power plants.

Local contributions to acid rain are produced by three types of combustion sources:

1. Combustion of residual oil in power plants produces both primary sulfates and sulfur oxides, which are discharged from stacks of intermediate height. Vanadium and nickel catalysts are also emitted.

2. Combustion of heating fuels (residual and distillate oil) in commercial installations and residences produces primary sulfate in a relatively higher proportion to SO_2 emissions than does the firing of residual oil in larger oil-fired boilers. These commercial/residential installations also produce sulfur oxides, which are discharged at low levels, and possibly some catalysts.

3. Combustion of automobile fuels in densely populated areas produces NO_x emissions that accumulate in the air masses near ground level. Sulfuric acid may also be emitted from catalytic pollution control devices.

The role of catalysis in acid rain production is recognized as very important, although it is not well understood. Catalysts accelerate what would ordinarily be very low atmospheric conversion rates of SO_2 and NO_x to SO_4= and NO_3-. In areas such as the Northeast, where large amounts of residual oils are burned, metal catalysts are emitted in large quantities. Vanadium, nickel, and other metal catalysts originally present in the fuel oil are later emitted in the flue gas. Once emitted, these metals can continue to operate as catalysts in local atmospheres rich in SO_2, NO_x, oxygen, and ozone. Therefore, it is reasonable to expect that these metals emitted by local oil-burning sources are acting together with ozone to catalyze the conversion of SO_2 and NO_x in local atmospheres. The reaction processes are further intensified by relatively high humidities and by the likely presence of other catalytic substances, such as iron, manganese, and carbon particles.

The plausibility of this scenario is supported in a report showing a correlation between ambient concentrations of vanadium and sulfates in 10 Japanese cities[2]. The report states that much of the ambient sulfate is formed with the help of a vanadium catalyst. It also presents data showing that high relative humidities favor the formation of sulfates.

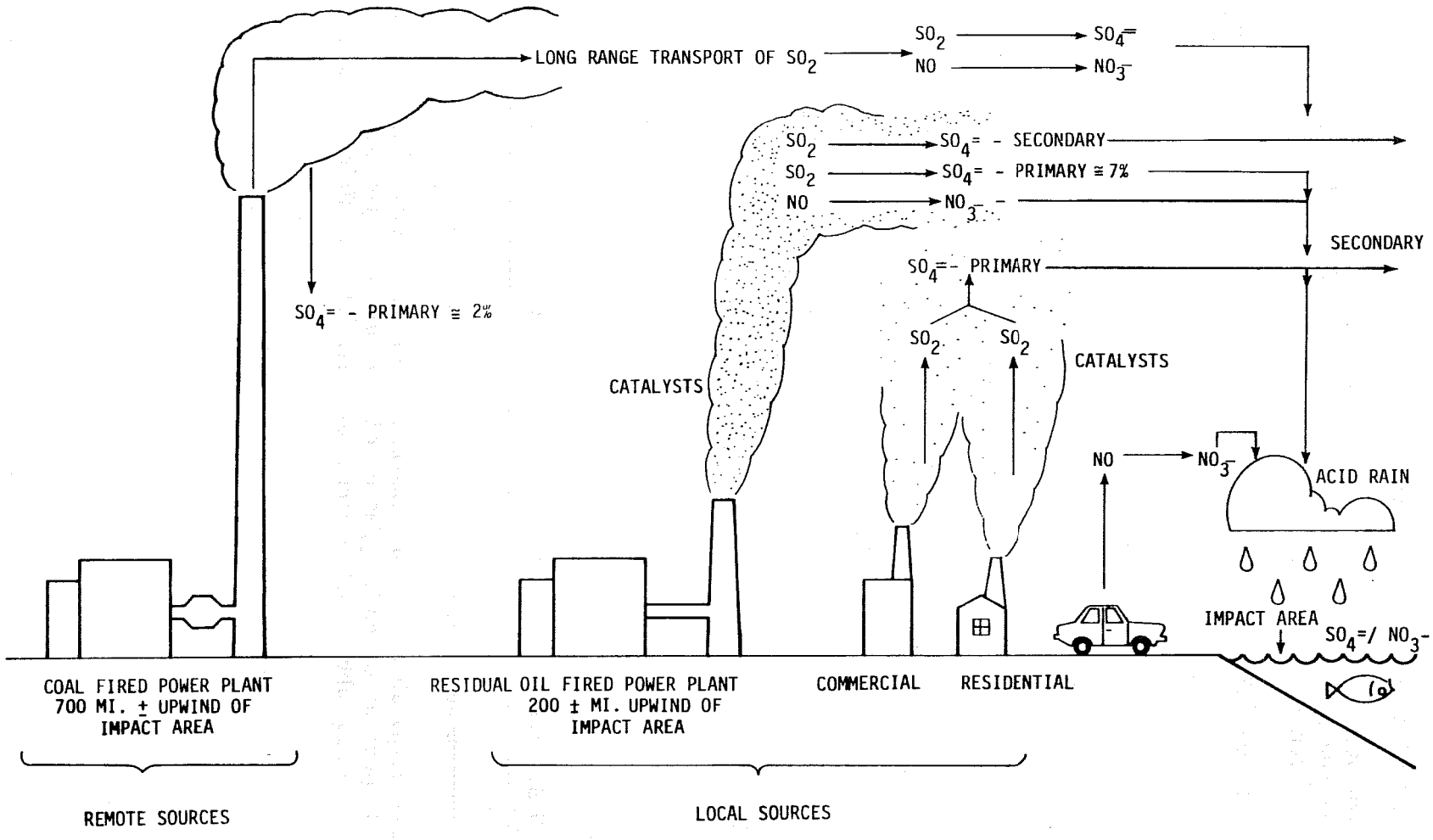


Figure 2. Simplified illustration of various contributors to the acid rain problem.

Oil Burning in Local Stationary Sources

Data on consumption of residual oil in the eight states contiguous with the impacted Adirondack areas are shown in Table 2, along with percentages of the total national consumption of residual oil.

TABLE 2. 1978 CONSUMPTION OF RESIDUAL OIL BY SECTOR IN STATIONARY SOURCES, 10^{12} BTU[a]

	Commercial	Industrial	Utility	Total
Connecticut	34	64	127	225
Massachusetts	109	34	305	448
New Hampshire	7	8	20	35
New Jersey	75	91	167	333
New York	337	62	520	919
Pennsylvania	60	129	162	351
Rhode Island	11	6	8	25
Vermont	4	3	0	7
Subtotal	637	397	1,309	2,343
U.S. Total	1,304	1,454	3,352	5,840
Percent of U.S. Total	61.6	27.3	39.1	40.1

[a]Source: Based on Reference 19.

Table 3 shows comparable data on consumption of distillate oil. The data show that about 35 percent of both types of oil is burned in the eight states whose land area is only about 4.5 percent of that of the 48 contiguous states.

TABLE 3. 1978 CONSUMPTION OF DISTILLATE OIL IN STATIONARY SOURCES BY SECTOR, 10^{12} BTU[a]

	United States	Eight States	Percentage of U.S. Total
Residential[b]	2,238	1,119	50.0
Commercial	1,117	445	39.8
Industrial	1,154	146	12.7
Utility	423	51	12.1
Total	4,932	1,761	35.7

[a]Source: Based on Reference 19.
[b]Includes distillate oil and kerosene.

Table 4 gives data on sulfur oxide emissions from oil burning sources; the data show that emissions in this area are about 1/3 of the national total.

TABLE 4. 1978 SO_x EMISSIONS FROM OIL FIRING IN STATIONARY SOURCES, 10^3 TONS[a]

	U.S.	Eight States	Percent of U.S. Total
Residual Oil	3,700.85	1,397.48	37.8
Distillate Oil	467.60	90.54	19.4
Total	4,168.45	1,488.02	35.7

[a]Values in Tables 4 and 5 based on Reference 19 and appropriate emission factors.

Sulfur oxide emissions associated with stationary sources in the eight northeastern states amount to 1.4×10^6 tons from residual oil consumption and 0.1×10^6 tons from distillate oil. The combined total of sulfur oxides from oil burning in the eight states is only 6 percent of the estimated 27×10^6 tons emitted annually from all sources in the United States. There is reason to believe, however, that emissions of sulfur oxides from local sources burning oil contribute to acid rain much more than would be expected on the basis of total annual emissions.

In the eight-state area, residual oil is burned mostly in power plants, but significant quantities are burned in commercial and industrial boilers. Utility boilers burning residual oil have been reported to produce 3 to 10 times more primary sulfate per unit of sulfur content than do those burning coal[20,21].

Industrial and commercial boilers, which are generally less efficient and more likely to use higher levels of excess air, are even more likely to produce sulfates directly. A recent study of 20 small distillate oil-fired boilers showed that an average of 15 percent of total emissions was sulfate.[22] Although the average sulfur content of distillate oil burned in the region is only about 0.2 percent, the mode of operation of combustors in commercial and residential facilities, where most of the distillate oil is burned, is such that direct sulfate production is very high. Estimates of the total emissions of primary sulfates are shown in Table 5.

TABLE 5. 1978 PRIMARY SULFATE EMISSIONS FROM OIL FIRING IN STATIONARY SOURCES, 10^3 TONS

	U.S.	Eight States	Percent of U.S. Total
Residual Oil[a]	252.74	95.46	37.8
Distillate Oil[b]	70.14	13.51	19.4
Total	322.88	108.97	33.7

[a]Based on 6.87 percent of SO_2 as primary SO_4=.
[b]Based on 15 percent of SO_2 as primary SO_4=.

In the area where acid rain appears to be most prevalent, the consumption of oil and production of sulfur oxides and sulfate are inordinately high. The data, however, provide only circumstantial evidence that local sources play a prominent role in production of acid rain. Therefore, a simple modeling exercise was conducted to develop perspective on the possible contribution of sulfates from local sources relative to secondary sulfates from remote coal-burning power plants. The results are discussed later in this section.

Consumption of Fuel for Local Transportation

Thirty percent of the acidity of rainfall in the Northeast is due to nitric acid[9]. Local NO_x emissions in the northeastern states rival those of the more distant midwestern states. Both stationary and mobile sources are the principal emitters of NO_x.

In the United States as a whole, and in the eight-state northeastern sector, transportation accounts for 47 and 46 percent of total NO_x emissions, respectively. If all NO_x sources contribute equally to the nitric acid in northeastern rainfall, one would conclude that about 14 percent of the total acidity of the rainfall results from transportation-related NO_x emissions and about 16 percent results from NO_x emissions from stationary sources. There is, however, some evidence to suggest that mobile sources may be more important than these data suggest.

In the Northeast a highly concentrated traffic corridor extends from Boston to New York and Philadelphia. In New Jersey, for example, gasoline vehicles are reported to account for more than 1/3 of the NO_x emissions. The entire state has been classified as a nonattainment area for photochemical oxidant and in

1977 had the highest NO_x density of any state[23]. This congested area lies south of the impacted areas of the Adirondacks western slopes. During most of the year, surface winds do not blow from this corridor toward the north. Normal surface winds arrive at the impacted area from Pennsylvania and other areas to the west. In summer, however, there is a definite change in the meteorology. Winds often originate in the south in the New York City area, passing through Albany before arriving at the western side of the Adirondacks[24].

The surface wind patterns suggest that high levels of transportation-related NO_x are directed toward the Adirondacks during the summer months and are converted to the acid forms either during transport or during the precipitation event.

Other evidence tending to associate motor vehicles with acid rain is shown in Figure 3 in which pH isopleths are superimposed on maps showing the air quality attainment status of the region of interest. Data on the eight states show widespread nonattainment of the standards for ozone, which may be associated with motor vehicle pollution. In contrast, no counties in the area of heaviest acid rain impact show nonattainment of SO_2 standards.

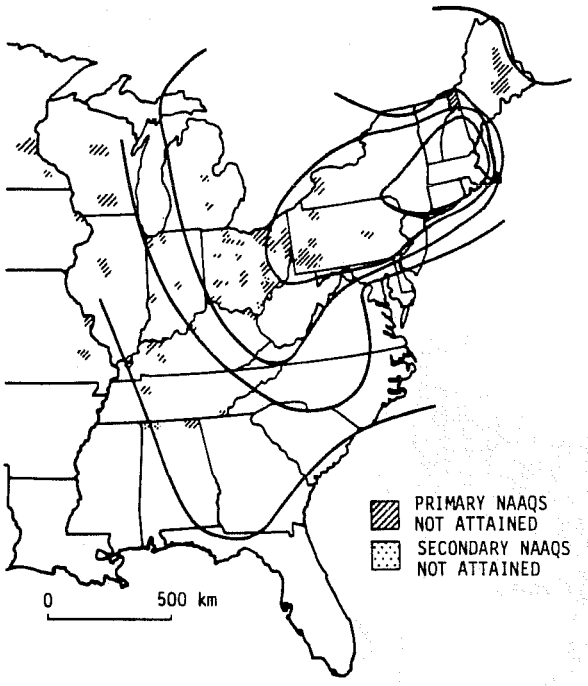
Mathematical Modeling

A simple mathematical box model was developed to indicate the percentages of sulfate in the northeastern United States that are attributable to local sources and to long-range transport and transformation of SO_2 from large, remote coal-fired power plants. The model represents four geographic areas, successively downwind from each other, in which atmospheric chemical reactions approximate those of a continuously-stirred tank reactor. The main variables that this model can handle are rate of transport, rate of transformation, rate of deposition, and background level of the air mass entering the first of the four areas for which sulfate deposition is estimated.

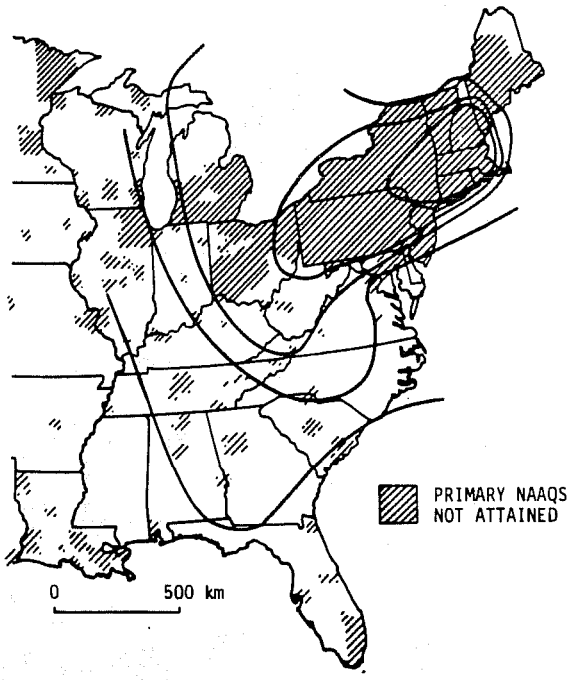
Use of this model showed that different assumptions relative to the major variables could affect the results, but that the emissions were more important in determining the contributions of local versus remote sources. For this exercise, the calculated sulfur oxide output from the 50 coal-burning power plants that produce the most sulfur oxide emissions (representing approximately 42 percent of total U.S. utility SO_2 emissions) was entered into the three regions or areas upwind from the impacted eight-state area. The power plants and areas are shown in Figure 4. For Area 4 (the impacted region) inputs were made for local oil- and coal-burning sources, and the relative sulfate deposition from local sources was compared with that estimated to be coming from the upwind top 50 coal-burning SO_2 emitters. Background levels were established for SO_2 and SO_4= and assumptions for rate of transformation and rate of deposition were varied to account for ranges of values given in the literature. These variations cause the contribution by local sources to range from 29 to 34 percent. If it is assumed that atmospheric catalysis causes the local deposition of 25 to 75 percent of the local SO_2 emissions as sulfates, the contribution of local sources ranges from 36 to 70 percent.

Admittedly the model is not sophisticated, amounting to little more than a material balance that shows under various assumptions, the amount of sulfate that might be deposited from remote and local sources. The results, however,

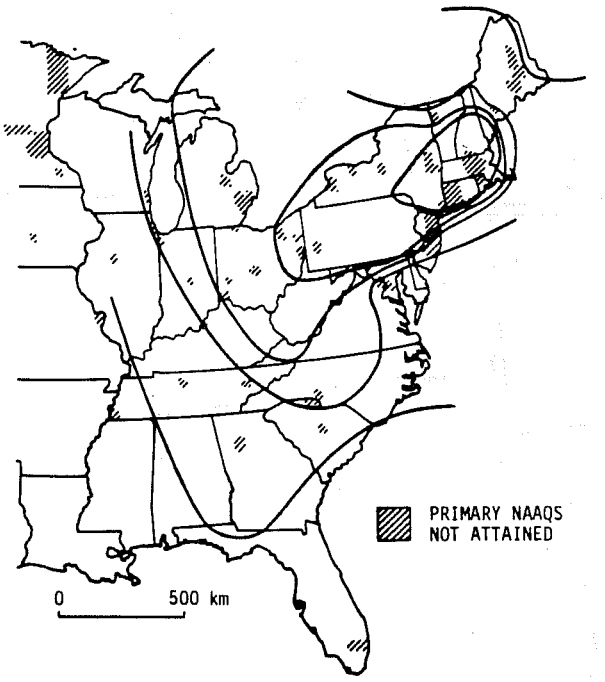
do point to the need for better definition of the role of local sources in formation of acid rain.



NON-ATTAINING COUNTIES FOR SO_2 - 1978



COUNTIES WITH OXIDANT CONCENTRATIONS GREATER THAN 0.08 ppm - 1978



NON-ATTAINING COUNTIES FOR CO - 1978

Figure 3[9,25]

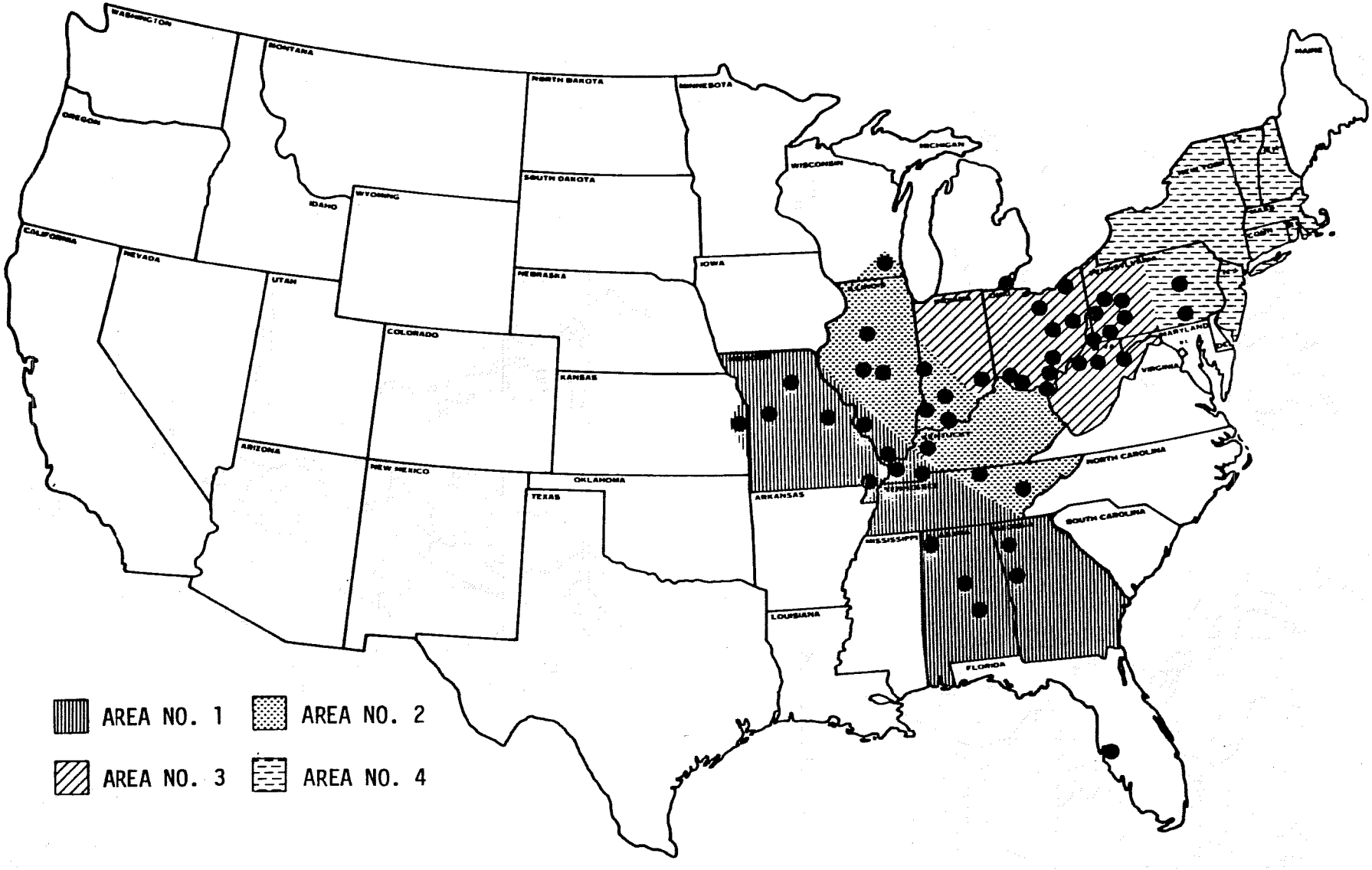


Figure 4. Illustration of 4 areas assumed for model and relative location of largest emitters of SO_2.

CAUSES OF ACID RAIN IN FLORIDA AND CALIFORNIA

As indicated earlier, acid rain has been observed in areas where its occurrence cannot reasonably be attributed to long-range transport of sulfates from coalburning power plants. Two such locations are northern Florida and California.

Florida

Researchers from the University of Florida at Gainesville report that rainfall in Florida has become markedly more acidic in the past 25 years. Brezonick et al.[13], have monitored rainfall at 24 stations throughout the state since 1977 at station locations shown in Figure 5. The studies have shown that the annual average pH value of rainfall in the northern three-quarters of the state is now below 4.7. The pH levels of summer rainfall are lower than those in winter. The authors report that over the past 25 years, the nitrate concentrations have increased by 450 percent and the sulfate concentrations have increased by 160 percent. Sulfuric acid is now 2 to 2.5 times as important as nitric acid in affecting the pH of precipitation.

Other researchers at Florida State University's Department of Oceanography (Tallahassee) report that rains in Tallahassee are more acidic when associated with air masses from the north[26]. Fourteen precipitation events occurring between November 7, 1978, and January 26, 1979, were studied. Air originating in more populated areas to the north produced rain with an average pH of 4.4 and a minimum of 3.7. Southern air flowing in from the gulf produced rainfall with an average pH of 5.3 and a minimum of 4.6. A strong correlation was observed between the log of the sulfur concentration and pH in northern rain, but not in southern rain.

It is not clear what is causing the acid rain in Florida. It may be partly attributable to long-range transport of pollutants from the neighboring states such as Georgia, Alabama, and Mississippi. It may, however, be partly attributable to anthropogenic activities within the state. Probable local sources include combustion of residual oil in power plants, transportation emissions, and sulfate from activities associated with the extensive phosphate-producing industry in Florida.

Florida has 56 power plants, of which 52 burn oil or oil and natural gas. Only four burn coal as the principal fuel[27]. Most plants are located in the northern two-thirds of the state, where acid rain is known to be occurring (see Figure 5b). In 1978, Florida consumed nearly 73 million barrels of residual oil, the third highest consumption among the 50 states. In consumption of all petroleum products (mainly residual oil and gasoline) Florida ranked sixth[19].

Utilities account for about two-thirds of Florida's total sulfur oxide emissions; phosphate production accounts for 10 to 15 percent. Mobile sources and utilities account for 85 percent of the NO_x emissions[28]. These figures indicate that oil-burning power plants and motor vehicles are the most likely local contributors to acid rain in Florida.

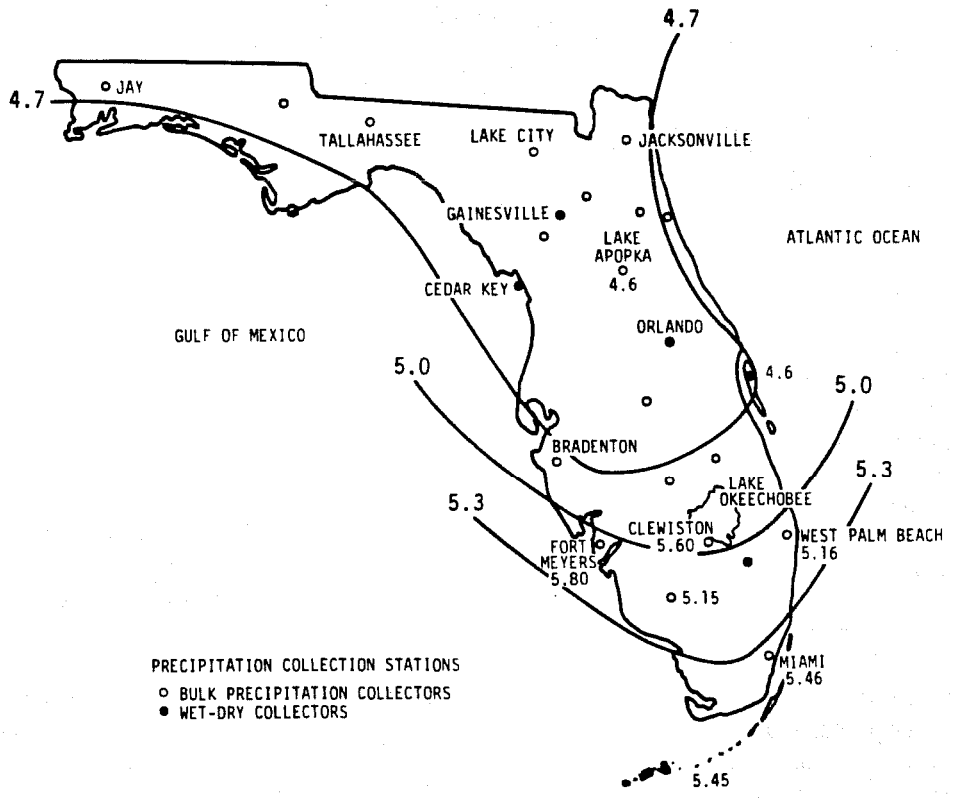


Figure 5a.

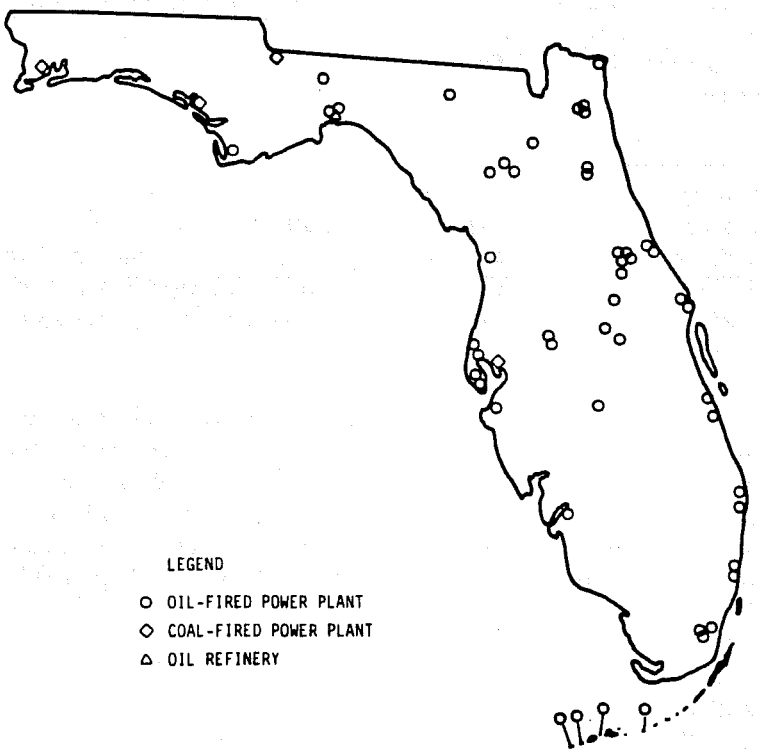


Figure 5b.

Figure 5a. Isopleths for pH of rainfall in Florida from May 1978 - April 1979, as reported by Brezonik, et al., 1980. Figure 5b. Locational map of refinery and utilities in Florida.

For states to the north and west of Florida (Alabama, Georgia, and Mississippi), approximately 80 percent of the sulfur oxide emissions are from utility and industrial sources. Nitrogen oxides are derived from both mobile and stationary sources in about equal amounts. Stationary sources in these states burn nearly equal amounts of coal and oil on an equivalent Btu basis[19]. It appears that much of residual oil is burned in the southern regions of these states, while coal burning is concentrated in the north. If acid rain in Florida is due in part to transport of pollutants from these upwind states, it may be attributable mainly to the automobile use and oil burning nearby with coal burning and long-range transport making some contribution.

California

Recent studies have shown that acid precipitation is occurring throughout much of California[8,14], as shown in Figure 6. In the northern part of the state, the mean pH of rainfall (1978-79) ranges from 4.4 to 5.3. Nitrates dominate sulfates at most northern locations with ratios* of NO_3^- to $SO_4^=$ ranging from 1.2 at Davis to 3.1 at Kearney. An exception is Tahoe City, where the ratio of NO_3^- to $SO_4^=$ is only 0.5, an indication that sulfates are predominant.

In southern California, the mean pH of rainfall (1978-79) ranges from 4.4 to 5.4[14]. The acidity pattern associated with the precipitation differs noticably from that of the northern region. At five of nine sampling locations (Westwood, Los Angeles, Long Beach, Pasadena, and Mt. Wilson) the ratios of NO_3^- to $SO_4^=$ are less than unity (range 0.4 to 0.9), an indication that sulfates are the largest contributor to rainfall acidity in those areas. All of the remaining sampling sites, where NO_3^- dominates $SO_4^=$, are inland locations, two of which are remote sites on the eastern side of the mountains surrounding the Los Angeles Basin. On the average, the NO_3^- levels in southern California precipitation are 62 percent higher than in the north; sulfate levels are 2.75 times higher.

The ratio of sulfuric acid and nitric acid derivatives in precipitation may be indicative of the predominant type of source contributing to acid rain in an area. A relatively high proportion of SO_x or sulfuric acid derivatives is thought to be indicative of stationary sources such as power plants, smelters, and heavy industry, whereas a relatively high proportion of NO_x or nitric acid derivatives is more suggestive of mobile sources.

The sulfate-related portion of rainfall acidity in California is not attributable to coal-burning utilities or smelters, since there are no such installations within the state. Local oil-burning utilities are a more likely source of a large part of the sulfate-related acidity. California is the second largest oil-consuming state, burning nearly 13 percent of all the residual oil consummed in the United States[19]. Oil-burning power plants and possibly oil refineries would be expected to emit not only large amounts of SO_x but also significant quantities of primary sulfates and vanadium catalyst. A review of the location of California's 41 oil-burning utilities and 41 oil refineries shows concentrations of 67 of these sources in three areas[27,29]:

* Ratios are based on equivalent weights.

LOCATION	MEAN pH
NORTHERN CALIFORNIA	
BERKLEY	4.66
MENLO PARK	5.30
DAVIS	5.20
SAN JOSE	4.42
HOPLAND	5.10
NAPA	4.84
CHALLENGE	4.88
TAHOE CITY	5.17
KEARNEY	4.96
Average	4.99
SOUTHERN CALIFORNIA	
WESTWOOD	4.53
LONG BEACH	4.54
LOS ANGELES	4.49
PASADENA	4.41
AZUSA	4.66
MT. WILSON	4.99
WRIGHTWOOD	4.90
RIVERSIDE	4.97
BIG BEAR LAKE	5.42
Average	4.76

Figure 6. California sites where pH of rainfall has been studied (1978-79). Source: McColl, 1980; Morgan and Liljestrand, 1980.

- The Berkeley-Oakland-Stockton area (13 sources)
- The Los Angeles-Long Beach area (42 sources)
- The Bakersfield area (12 sources)

Because acid rain is known to be occurring in two of the three areas (Bakersfield has not been monitored), and because upwind, long-range sources are not a factor in the San Francisco or Los Angeles areas, it is reasonable to suspect that emissions of SO_4= from local oil-burning/oil-producing sources are contributing to the acidity of precipitation in nearby areas.

McColl believes that the high ratios of NO_3- to SO_4= in northern California are due to NO_x emissions from automobiles[8]. Estimates for total NO_x for the state as a whole tend to support this view, NO_x from motor vehicles being over twice the amount from stationary sources.

To summarize, while these data are only suggestive as far as contributing sources in California are concerned, it seems quite conclusive that coal burning and long-range transport are not a factor in California.

5. IMPLICATIONS OF THE FINDINGS

Long-range transport of SO_2 and its transformation to SO_4= have been much studied, along with the possible contribution of remote sources of SO_2 to acid rain in the northeastern United States. The magnitude of impacts from such sources, relative to contributions from other sources of SO_2 and NO_x, has not been quantified.

The role of local sources in production of acid rain in the northeastern United States, which is believed to be heavily impacted, has been given little consideration despite evidence from other parts of the United States suggesting that local sources may contribute to acidification of rain and surface waters in the surrounding areas.

Examination of the potential contribution of combustion of petroleum products (residual oil and gasoline) to the acid rain in California and Florida suggests, in the absence of other possible major contributors, local or remote, that oil-burning power plants and automobiles are the most likely causes.

Examination of data on the burning of oil (distillate and residual) and on consumption of transportation fuels in the northeast United States indicates that local oil burning and consumption of gasoline in automobiles may be a significant, and perhaps dominant, cause of acid rain in that region.

Local contributions of nitrogen oxide from automobiles and small combustion units, as well as primary sulfates from the combustion of oil, are emitted at ground level with much less opportunity for dispersion as compared to distant sources. This implies a potentially more severe local concentration of acid rain precursors. These high local concentrations could have increased material damage effects since materials damage would be expected to be somewhat proportional to concentration of acid rain. Strategies aimed at reducing distant sources could have only minor effects on the concentration of local pollutants and provide minor relief for material damage caused by acid rain and acid rain precursors.

Currently, the Federal Government is considering the costs associated with different strategies intended to control acid rain. The focus of the analyses performed to date is on coal burning power plants that were constructed prior to implementation of the Clean Air Act. These strageties would have profound economic implication and may not achieve the desired results. For example, application of flue gas desulfurization to the 50 largest coal-fired SO_2 emitters east of the Mississippi would cost in the neighborhood of $10 to $15 billion. Other alternatives that have lower direct costs, for example, a shift to low sulfur coal, could place significant socioeconomic burdens in major regions within the nation.

The prospect of needing to consider the role of local acid rain precursors suggests the need to reconsider the environmental effects of replacing oil with coal as a energy source. Combustion of oil generates a significantly different set of pollutants such as potentially toxic trace metals and partially burned hydrocarbons in fine particulate form, as well as the classical sulfur oxide and nitrogen oxide. Many authorities feel these oil combustion

pollutants may be more hazardous than equal concentrations of coal combustion pollutants. If so, switching from oil to properly controlled coal combustion systems might have benefits beyond the national objective of reducing oil imports.

The conclusions to date relative to the role of local sources in production of acid rain are based on a modest amount of evidence. All signs point, however, to a need for further investigation of local sources, especially those utilizing petroleum products, to determine their importance relative to that of remote coal-burning power plants in production of acid rain. Work in this connection is continuing and is expected to produce more definitive conclusions in the near future.

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