

# Appendix C

## Atmospheric Processes

### C.1 ATMOSPHERIC CHEMISTRY (p. 265)

### C.2 SOURCE-RECEPTOR RELATIONSHIPS (p. 274)

## C.1 ATMOSPHERIC CHEMISTRY\*

The atmosphere is a mixture of chemicals—some of natural origin, some anthropogenically generated, and some that are produced by nature as well as by man. These chemicals can react with each other to varying degrees under varying conditions. Because the atmosphere is dynamic, the particular chemical environment it represents is different for every location, every season, and every meteorological condition.

Both acid deposition and ozone are produced by transported air pollutants. The dominant precursors of acid deposition are sulfur dioxide ( $\text{SO}_2$ ) and nitrogen oxides ( $\text{NO}_x$ ). Ozone formation involves  $\text{NO}_x$  and reactive hydrocarbons (RHC). Anthropogenic sulfur oxides ( $\text{SO}_x$ ) are produced primarily by burning such sulfur-containing fossil fuels as coal and oil. Nitrogen oxides also result from the burning of fossil fuel by utility, industrial, and mobile sources. Anthropogenic hydrocarbon emissions result primarily from petroleum refining and storage, other industrial process emissions, and mobile sources.

After release into the atmosphere, the particular sequence of changes a pollutant undergoes depends on the physical and chemical characteristics of the air mass in which it travels. Ultimately, though, because the atmosphere is 20 percent oxygen, emitted  $\text{SO}_x$  and  $\text{NO}_x$  will oxidize\*\* and can form acid when combined with water. This can occur while the pollutants are in the air, or following deposition on the Earth. The acids formed may subsequently be neutralized if appropriate chemical species are available.

Which of many chemical routes is actually followed depends on: 1) the initial concentrations of all pollutants; and 2) a number of physical factors, such as wind speed, air turbulence, sunlight intensity, temperature, and rainfall frequency.

While scientists know a great deal about individual

chemical reactions and specific physical processes, they cannot precisely characterize the detailed path of a pollutant from its origin or 'source' to its removal or 'sink. Reactions can occur when pollutants exist as gases, are dissolved in liquids, or adhere to particles. In general, gas-phase reactions predominate in the transformation of  $\text{NO}_x$  and the production of ozone, while all three phases—gas, liquids, and solids—are presently believed to play a part in  $\text{SO}_x$  transformations under different atmospheric conditions. Nonetheless, the key atmospheric reactions involved in producing acid deposition and ozone have some common features and are closely interrelated.

Energy from sunlight triggers chemical reactions that transform  $\text{SO}_x$  and  $\text{NO}_x$  (which includes  $\text{NO}$  and  $\text{NO}_2$ ) into sulfuric and nitric acid, respectively. Such 'photochemical' processes, which also form ozone, require the presence of RHC. For example, hydroxyl ( $\text{OH}$ ) radicals—a very reactive chemical species—initiate the gas phase transformation of  $\text{SO}_2$  to sulfuric acid. Since concentrations of  $\text{OH}$  depend, in turn, on concentrations of ozone,  $\text{NO}_x$ , and RHC, as well as on sunlight intensity, the atmospheric chemistry of ozone,  $\text{NO}_x$ ,  $\text{SO}_2$ , and RHC must be considered together. Altering the concentrations of any one of these will affect pollutant transformation and deposition rates.

### Deposition of Sulfur and Nitrogen

In the East, approximately two-thirds of the acid deposited results from sulfur compounds, and one-third results from nitrogen compounds. Over much of the West,  $\text{NO}_x$  emissions play a relatively greater role in acidification than in the East. To become acids, emitted  $\text{SO}_2$  and  $\text{NO}_x$  must be oxidized either: 1) in the gas phase, 2) after absorption into water droplets, or 3) after dry deposition on the ground.

These materials can be deposited on the ground unchanged (as primary gaseous pollutants), or in a *transformed* state (as secondary pollutants). Transformed pollutants can be deposited in *wet* form (as rain, fog, or snow), or *dry* form (due to particles containing these materials settling out). The amount of time a pollutant remains in the atmosphere, and therefore, how far it

\* This appendix is based on the OTA contractor report, "An Assessment of the Atmospheric Chemistry of Oxides of Sulfur and Nitrogen Acid Deposition and Ozone," by B. J. Finlayson-Pitts and J. N. Pitts, Jr., 1982.

\*\* Oxidation is the process of adding oxygen to a chemical. Oxidizing agents or 'oxidants' are chemicals that supply the extra oxygen needed to convert  $\text{SO}_2$  to sulfate, or  $\text{NO}_x$  to nitrate. When sulfate and nitrate combine with water, sulfuric acid and nitric acid are formed unless neutralized by other chemicals present.

is transported, depends significantly on its chemical form. For example,  $\text{SO}_2$  gas is dry-deposited at a greater rate than sulfate particles (products of oxidation). If  $\text{SO}_2$  is quickly converted to sulfate, a smaller fraction of emitted sulfur will be deposited locally, in the absence of precipitation. The rate of conversion of  $\text{SO}_2$  to sulfate depends on the chemical composition of the atmosphere. The frequency and intensity of precipitation controls the rate of wet sulfur deposition.

Dry deposition is believed to occur at a fairly constant rate over time (i. e., a certain percentage of the  $\text{SO}_2$  in the air is dry-deposited each hour), but varies somewhat over different terrain. Wet deposition is episodic, and the amount deposited varies considerably even within a rainfall event. For example, a short rain may deposit heavy doses if pollutants are concentrated—e.g., if they have been forming and accumulating in the air over time. Without sufficient time for pollutant concentrations to accumulate and be transformed, a second rain in quick succession may deposit little new acid material. The product of concentration times rainfall determines the total dose of wet-deposited acidic material.

For this reason, acid fogs (recently identified in the Los Angeles basin) may expose the area to a very high concentration of acid (a large quantity of acid per amount of water), but significantly smaller amounts of acid may be deposited than would occur from a rainfall. On an annual basis, the contribution to total acid loadings depends on the percent of time that fog covers an area. Particularly in high-altitude regions (e. g., the Northern Appalachian mountains) where cloud cover and fog persist for about 50 percent of the time, precipitation may account for less acid deposition than fog. The relative effect of high acid concentrations (but low total doses) as compared to high total doses of acid (at low concentrations) depends on the nature of the receptor or ecosystem in question. (See app. B, "Aquatic Resources, Terrestrial Ecosystems, and Materials.")

In general, areas close to emission sources receive significant proportions of their pollution from steady dry deposition of  $\text{SO}_2$ . In locales remote from emission regions, much of the  $\text{SO}_2$  available for dry deposition has been depleted or converted to secondary pollutants. In these areas, wet deposition delivers a greater share of the total pollutant dose than does dry deposition (see fig. C-1). Over most of the nonremote Eastern United States, the contribution from wet and dry deposition is estimated to be about equal. Air over any particular area will carry some residual pollution from distant areas, as well as infusions received from more recently passed areas. The continued replenishment and depletion of pollutants along the path of an air mass makes precise source-receptor relationships extremely difficult to determine.

### Atmospheric Chemistry of the Oxides of Sulfur

About 26 million tons of manmade  $\text{SO}_2$  are emitted in the continental United States. About 22 million of these tons are emitted in the Eastern 31-State region. The oxidation of natural sulfur compounds could contribute significantly to atmospheric  $\text{SO}_2$  concentrations in regions where natural emissions are high (e. g., from volcanoes, or some types of marshes) and manmade emissions are low. However, on a nationwide basis, less than 5 to 10 percent of sulfur emissions are attributed to natural sources.

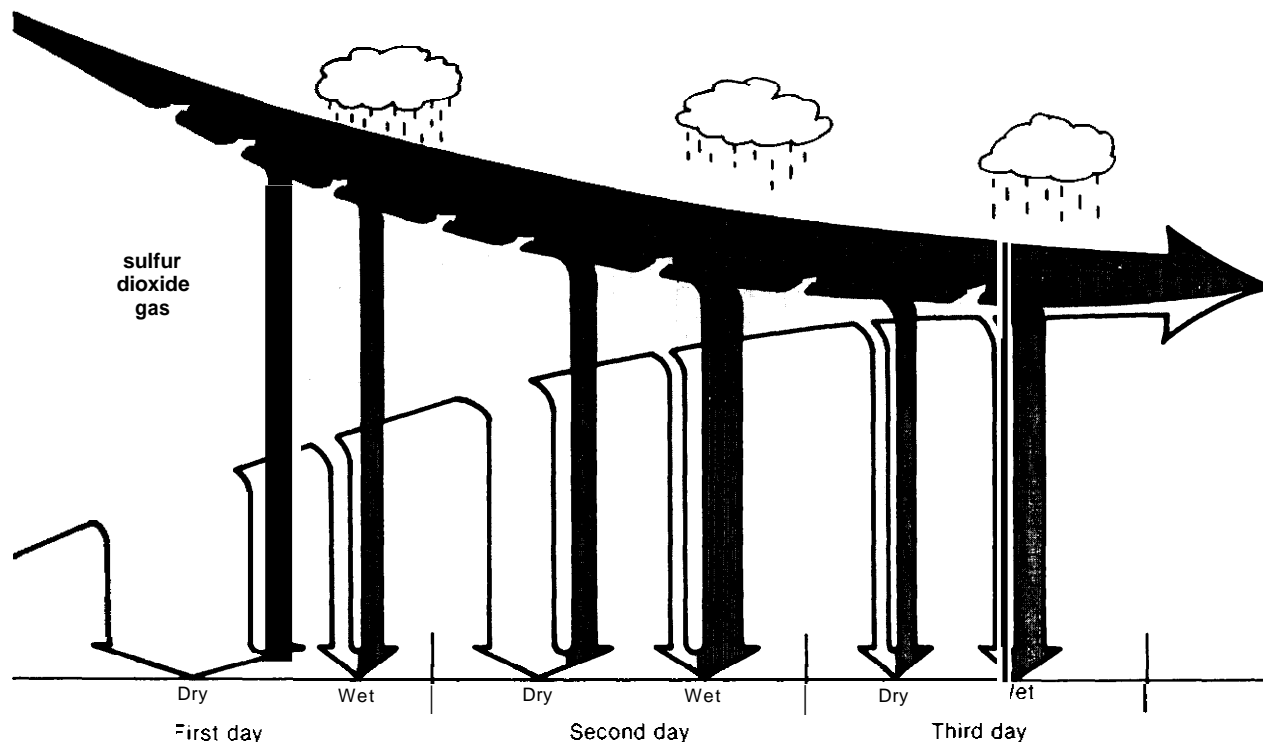
The following discussion of the various fates of emitted  $\text{SO}_2$  is summarized in figure C-2. One way in which sulfate is formed involves  $\text{SO}_2$  gas interacting with OH radicals in a *homogeneous gas phase reaction*—i. e., the reactants are all in the gas phase. Because OH is highly reactive with many atmospheric components, each OH radical has a short lifetime in the atmosphere. Sunlight is necessary for triggering the chain reaction leading to OH production. Consequently, the greatest quantity of  $\text{SO}_2$  gas is oxidized by OH radicals during periods of intense sunlight—i. e., at midday, and in the summer. The maximum rate at which this reaction converts  $\text{SO}_2$  to sulfate is estimated to be about 1 to 4 percent per hour. However, field experiments show conversion rates significantly greater (10 to 30 percent per hour) than homogeneous gas-phase reaction rates. Therefore, significant quantities of sulfate must be produced by aqueous (liquid) phase reactions or heterogeneous reactions involving two phases (i. e., reactions of gases on either liquid droplets or solid particles).

In *aqueous phase reactions*,  $\text{SO}_2$  is dissolved in water droplets, where oxidants convert the  $\text{SO}_2$  to sulfate. There is little agreement as to which oxidizing agent (the candidates include dissolved oxygen, ozone, metals, hydrogen peroxide, free radicals, and  $\text{NO}_x$ ) is most important under particular conditions. The rate of each oxidizing process may depend on the acidity of the solution; the relative importance of particular oxidizing agents may, therefore, change as acid is formed and the pH\* of the water droplet decreases. As acidity increases,  $\text{SO}_2$  is also less easily dissolved, which slows down some aqueous phase reactions significantly. (For example, the presence of nitrate compounds can increase the acidity of droplets, allowing less  $\text{SO}_2$  to dissolve; the presence of ammonia in the atmosphere can buffer such increases in acidity, allowing more  $\text{SO}_2$  to dissolve.) Current research suggests that the major aqueous phase oxidation route for  $\text{SO}_2$  under typical ambient conditions is

\*pH is related to acidity. Decreasing pH corresponds to increasing acid. The pH scale is not linear; a drop of one pH unit reflects a tenfold increase in acidity. Compared to a pH of 7 (neutral), a solution of pH 6 is 10 times more acid, pH 5 is 100 times more acid, and pH 4 is 1,000 times more acid.

**Figure C-1.—The Effects of Time and Distance on Conversion and Deposition of Sulfur Pollution**

Sulfur can be deposited in both its emitted form, sulfur dioxide (lighter shading), and as sulfate, after being chemically transformed in the atmosphere (darker shading). Both compounds can be deposited in either dry or wet form. The relative amount of sulfur deposited in these forms varies with distance from emission sources. Dry deposition predominates in areas close to emission sources. Wet deposition is responsible for a larger percentage of pollutant load in areas distant from source regions.



SOURCE: Office of Technology Assessment

the reaction with the oxidant hydrogen peroxide, because this reaction occurs quickly and appears to be relatively independent of pH.

A variety of measurements indicate that  $\text{SO}_2$  gas can be adsorbed onto particles (e. g., carbon soot in plumes) and then oxidized to sulfate. These types of *heterogeneous reaction* rates may be particularly significant in urban plumes.

Gas phase, liquid phase, and heterogeneous reactions may all be important under differing atmospheric conditions. For example, if there is no condensed water and the concentration of particulate surfaces is low, gas phase oxidation will predominate during daylight hours. However, if clouds or fog are present, oxidation in the aqueous phase can predominate. In either case, heterogeneous reactions on surfaces may also be important if sufficient surfaces associated with particulate are available. Such conditions are most likely near an emissions source—e.g., in a powerplant plume.

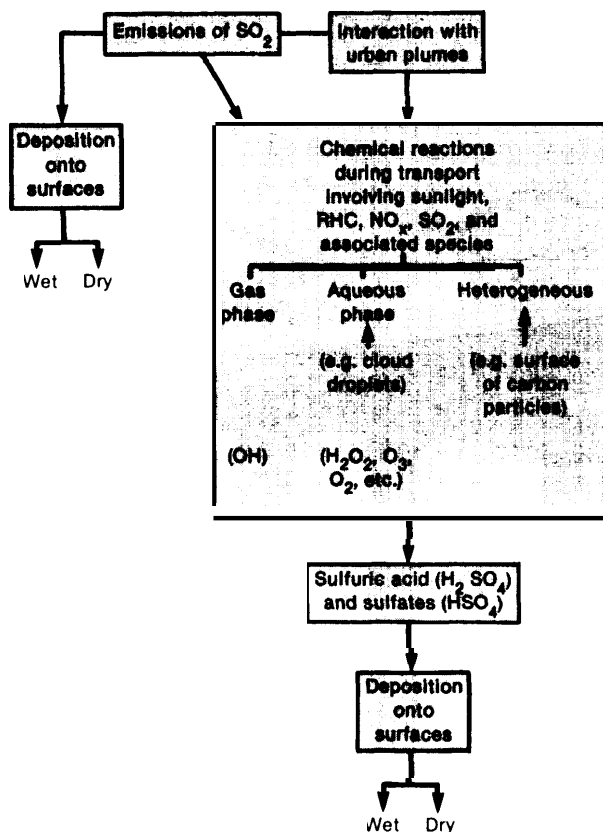
Overall, current estimates based on empirical observation and model results suggest that homogeneous gas-

phase reactions account for 25 to 50 percent of sulfate formed, and aqueous phase reactions account for the remaining 50 to 75 percent on a regional scale.  $\text{SO}_x$  can be converted to sulfate quickly in the aqueous phase or in concentrated plumes (e. g., more than 10 percent per hour) or slowly (e. g., less than 1 percent per hour in the dry winter) in the gas phase. Thus,  $\text{SO}_x$  are available for atmospheric transport for periods of 1 day to about a week, and will travel varying distances depending on meteorology and precipitation frequency. The form in which it is deposited depends on the chemistry described above. Ultimately, dry-deposited  $\text{SO}_2$  and sulfate may also produce acid on the surface of the Earth following oxidation and combination with available water (e. g., dew).

#### Atmospheric Chemistry of Oxides of Nitrogen

Manmade  $\text{NO}_x$  emissions result both from nitrogen bound in fuels and from compounds formed from nitrogen and oxygen in the air during combustion. Anthro-

Figure C-2.—Schematic Diagram of Possible Fates of Emitted Sulfur Dioxide Gas



SOURCE: Office of Technology Assessment, modified from Finlayson-Pitts and Pitts, OTA contractor report, 1982.

pogenic sources emitted about 21 million tons of  $\text{NO}_x$  in the continental United States during 1980. About 14 million tons were emitted in the Eastern 31-State region. Additionally, natural sources produce several forms of nitrogen compounds, primarily from soil processes, organic decay, and lightning. Natural sources of reactive  $\text{NO}_x$  emissions are estimated to produce less than 15 percent of total  $\text{NO}_x$  emitted in the Eastern United States. In North America as a whole, the natural background of  $\text{NO}_x$  may represent 5 to 35 percent of the total  $\text{NO}_x$  produced.

Gas phase reactions of  $\text{NO}_x$  have been studied for a number of years because of their role in forming ozone in photochemical smog. The importance of  $\text{NO}_x$  as a source of acid has only been investigated more recently.

Both ozone and nitric acid formation involve RHCs. The homogeneous gas phase reaction leading to nitric acid (involving the OH radical) occurs about 10 times as rapidly as the corresponding reaction with  $\text{SO}_2$ . Consequently, nitric acid formation and deposition should occur at distances closer to the source, constituting more

of a local phenomenon than sulfuric acid deposition. A recent modeling study of a very polluted area, the Los Angeles basin, suggests that 40 percent of emitted  $\text{NO}_x$  are transformed into nitric acid over the course of 1 day. Similar rapid conversion occurs in plumes from the northeast moving over the Atlantic Ocean, a much less polluted area.

In addition to nitric acid, other nitrogen-containing species are formed in polluted atmospheres—e.g., peroxyacetyl nitrate (PAN), a nitrogen-containing chemical known to be toxic to plants. In the winter, when PAN is less likely to be decomposed by heat, it may serve as a reservoir for  $\text{NO}_x$ , allowing substantial transport before either decomposition or deposition occurs.

Recent experimental evidence indicates that uncatalyzed aqueous phase reactions of  $\text{NO}_x$  compounds are too slow to be important under most atmospheric conditions. However, catalyzed aqueous reactions (e.g., due to the presence of metals or surfaces) may proceed quickly and therefore be important.

### Ozone

Ozone is regulated by National Ambient Air Quality Standards under the current Clean Air Act, and has been the focus of most oxidant control studies.  $\text{NO}_x$  react with sunlight and hydrocarbon-produced radicals to form ozone. Because ozone as well as its precursors can travel substantial distances, ozone concentrations downwind of emissions sources commonly exceed natural background levels.

Naturally produced hydrocarbons, including terpenes from pine trees, and methane from termites and wetland areas, can play a role in forming ozone. Also, additions of ozone from the upper atmosphere (the stratosphere) will contribute directly to observed levels. Such natural contributions can result in ozone concentrations on the order of 10 to 50 parts per billion (ppb), with common background measurements of about 20 to 30 ppb. At midlatitudes in the summer, natural ozone is augmented by photochemical ozone produced when pollutant precursors react with sunlight. (See app. B, "Terrestrial Resources at Risk From Ozone and Acid Deposition, for seasonal ozone levels throughout the United States.) Recent evidence<sup>2</sup> suggests that air masses with very low concentrations of  $\text{NO}_x$  and RHC (by air quality standards) produce high ozone values in the mountains of Colorado—up to or exceeding the ozone standard. Such concentrations may be quite common in the rural Midwestern and Eastern United States; high ozone levels may be produced at these sites, especially during the summer.

<sup>2</sup>F. C. Fehsenfeld, M. J. Ballinger, S. C. Liu, et al., "A Study of Ozone in the Colorado Mountains," *Journal of Atmospheric Chemistry*, in press

Some of the secondary pollutants (e. g., nitrous acid and formaldehyde) formed along with ozone can themselves facilitate further ozone production. These secondary pollutants can remain in a stagnant air mass overnight and react in the presence of sunlight the next morning.

#### IMPLICATIONS FOR CONTROL STRATEGIES

A nonreactive primary pollutant such as carbon monoxide (CO) emitted in an urban atmosphere does not, on the time scale of 1 day, undergo significant chemical reactions during diffusion, dispersion, and transport processes. For this pollutant, reducing emissions by 50 percent will reduce CO concentrations in ambient air by 50 percent. Thus, in California's South Coast Air Basin, although the number of vehicle miles traveled has increased substantially over the last decade, exhaust emission controls put on light-duty vehicles have reduced ambient CO levels.

However, primary pollutant emissions that can react relatively rapidly in the air to form secondary pollutants present an entirely different situation. Oxides of nitrogen and RHCs react in the presence of sunlight to form ozone and a host of other secondary pollutants such as formaldehyde, PAN, hydrogen peroxide, nitrous acid, and nitric acid, along with respirable particles. In such a complex system, the effect of reducing emissions is not as easy to determine as in the case of CO, i.e., it may not be "linear."

The term "linear" is used in connection with acid deposition to indicate that a given percentage reduction (or increase) in emissions would cause the same percentage change in acid deposition at a specific location. However, a number of factors may cause emissions reductions (of  $\text{SO}_x$ ,  $\text{NO}_x$ , or RHC) at one location to result in reductions in acid deposition that are *not* directly proportional at a particular site downwind. For example, introducing fresh supplies of  $\text{NO}_x$  and RHC (either individually or together) between controlled sources and a receptor could increase or decrease quantities of acid (both nitric acid and sulfuric acid) deposited at that site. In addition, the relative rates at which the various chemicals are deposited (e. g.,  $\text{SO}_2$  v. sulfate) may also influence the effect of emissions reductions on acid deposition at a particular location.

One of two possible fates occurs to sulfur and nitrogen pollutants: 1) oxidation to sulfuric and nitric acids (or sulfates or nitrates), followed by deposition at the Earth's surface; or 2) deposition in their emitted form (i. e., unchanged). In the latter case, chemical oxidation on the Earth's surface may cause the same net result—i.e., acidification.

Elevated concentrations of oxidizing chemicals such as ozone and hydrogen peroxide are found in atmos-

pheres containing anthropogenic emissions. However, it is possible that at certain times and places, the amount of  $\text{SO}_2$  or  $\text{NO}_x$  may exceed the supply of available oxidants in the atmosphere; when the supply is exhausted, additional acid cannot be made in the air until more oxidants become available. This would tend to delay acid deposition until the pollutants travel further downwind. Such conditions of "saturation" are thought to be episodic, and can generally be discounted in regional descriptions of transport and deposition.

Until recently, it had been assumed that "natural" water would have an acidity level, or pH, of about 5.6 (a pH of 7.0 being neutral, 0 to 7 being acidic, and 7 to 14 basic), due to atmospheric carbon dioxide dissolved in it. However, even "clean" atmospheres contain a host of other chemical constituents as well, which can interact to produce solutions more or less acidic than a pH of 5.6. For example, it has been shown recently that rainwater pHs of less than 5 might be expected in "clean air" environments under certain conditions when naturally produced sulfur and chlorine compounds are present (e. g., in coastal environments).

In determining whether the oxidation of  $\text{SO}_x$  and  $\text{NO}_x$  will actually cause acids to be deposited, the availability of neutralizing species such as ammonia and calcium (e. g., from windblown dust) must also be considered. Sufficient quantities of neutralizing species will reduce the actual acid formed in the atmosphere. Thus, while two sites may be subjected to identical concentrations of anthropogenically produced sulfate and nitrate, quite different levels of acid may be deposited if significant concentrations of neutralizing species are available at one location, but not at the other.

The ultimate effect of the deposited compounds, however, is of prime concern. Nonacidic sulfates are probably less damaging to *materials* than acid sulfates. However, these compounds could eventually act to acidify a *natural ecosystem*. For example, if ammonia combines with sulfates, the deposition will not be acidic. Yet ammonia, when used by plants and micro-organisms in soils, produces acidity, which can subsequently affect soils, lakes, and streams.

#### Altering Emissions of Primary Pollutants

Characterizing the "chemical soup" of the atmosphere at any specific location requires integrated information on the concentrations of all pollutants, the availability of oxidants, the predominance of gas or aqueous phase chemistry, and detailed meteorological information. The complexities involved in the various chemical and physical processes allow OTA to make only a very general description of the effect of changing emissions of various pollutants. The discussion refers to regional-

level response over time, and should not be interpreted as applying to particular episodic events or specific receptor sites.

#### REDUCING REACTIVE HYDROCARBON EMISSIONS

If  $\text{NO}_x$  concentrations remain at a freed level, reducing RHC emissions subsequently reduces ozone concentrations, by decreasing production of the free radicals necessary for ozone production. The peak ambient concentrations of ozone are then expected to be lower.

The only mechanism for producing *nitric acid* believed to be significant in urban areas and powerplant plumes is the oxidation of  $\text{NO}_2$  by the hydroxyl radical (and possibly by ozone) to produce other oxides of nitrogen which then form nitric acid. Reducing concentrations of RHC would lower the concentrations of the free radicals as well as of ozone; this should slow down the rate at which  $\text{NO}_x$  is oxidized to nitric acid. This slowing could in turn cause nitric acid formation and deposition over a larger geographical area, but at lower concentrations.

Reducing RHC emissions will similarly reduce the rate of gas phase oxidation of  $\text{SO}_2$ . The rate of liquid phase oxidation by hydrogen peroxide may also decrease because its formation rate is proportional to the concentration of OH radicals. The effect on other liquid phase transformations of  $\text{SO}_2$  are difficult to assess but are currently thought to be insignificant.

#### REDUCING NITROGEN OXIDES EMISSIONS

The question of how decreasing  $\text{NO}_x$  emissions while keeping RHC emissions constant would affect ozone levels is much more controversial than questions associated with the effects of RHC control. This is because the effect of reducing  $\text{NO}_x$  emissions depends on the resulting *ratio* of hydrocarbons to  $\text{NO}_x$ . Figure C-3 shows how ozone levels change with varying levels of  $\text{NO}_x$  and hydrocarbon emissions. Thus, when the ratio of RHC/ $\text{NO}_x$  is greater than indicated by the diagonal line in figure C-3 (i.e., for the right lower portion of the diagram), reducing  $\text{NO}_x$  emissions reduces ozone concentrations. However, at lesser ratios (i. e., for the upper left of the diagram), reducing  $\text{NO}_x$  emissions while holding RHC levels constant, leads to increases in ozone concentrations. \*

Figure C-3 implies that in the vicinity of emissions centers like downtown Los Angeles, where the RHC/ $\text{NO}_x$  ratio is less than that of the diagonal line, reducing  $\text{NO}_x$  emissions might increase ozone concentrations slightly. However, over an entire air basin, including regions several hundred kilometers downwind, reduc-

ing  $\text{NO}_x$  emissions should decrease ozone concentrations overall. Since  $\text{NO}_2$  is the sole precursor to anthropogenic ozone, this must hold true over a large region.

Because  $\text{NO}_x$  concentrations affect the availability of hydroxyl radicals, decreasing  $\text{NO}_x$  emissions may change the oxidation rate of  $\text{SO}_2$ . The change probably also depends on the RHC/ $\text{NO}_x$  ratio, but is poorly understood at present. However, reducing  $\text{NO}_x$  concentrations, regardless of the ambient hydrocarbon concentration, decreases the  $\text{NO}_x$  available for nitric acid formation and deposition.

#### REDUCING REACTIVE HYDROCARBON AND NITROGEN OXIDES EMISSIONS SIMULTANEOUSLY

Smog chamber and modeling studies (fig. C-3) show that simultaneous control of both RHC and  $\text{NO}_x$ —keeping their concentration ratio constant—would reduce ozone concentrations. As mentioned above, less nitric acid formation is also expected since the reduced  $\text{NO}_x$  limits how much acid can be formed. In this case, the oxidation rate of  $\text{SO}_2$  is also likely to be reduced.

#### REDUCING SULFUR DIOXIDE EMISSIONS (ALONE)

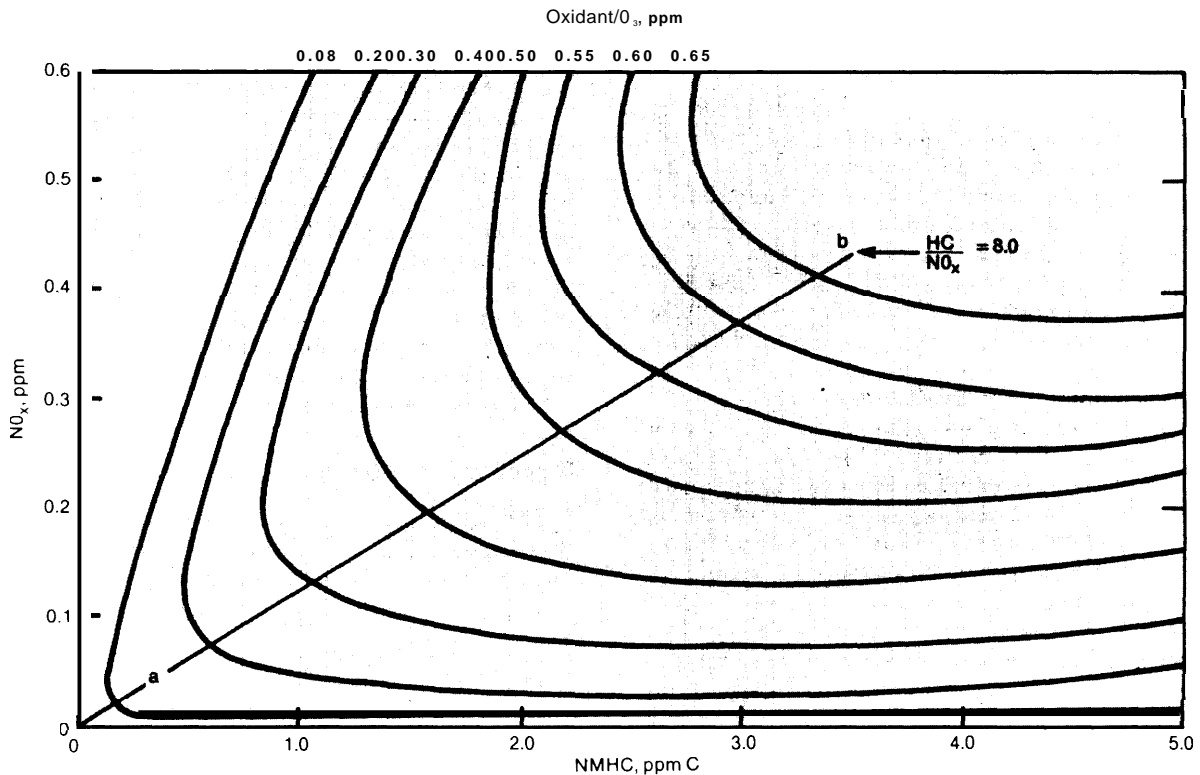
Available knowledge of atmospheric chemistry suggests that if: 1) there is no shortage of oxidizing agents (i.e., saturated conditions do not prevail); 2) levels of RHC and  $\text{NO}_x$  remain constant; and 3) deposition processes and meteorology remain constant, reducing  $\text{SO}_2$  emissions will reduce the total amount of acid formed in the atmosphere. Furthermore, given these conditions, a decrease in  $\text{SO}_2$  emissions should decrease the atmospheric formation of sulfuric acid by an approximately equal proportion.

#### REDUCING SULFUR DIOXIDE EMISSIONS CONCURRENTLY WITH REACTIVE HYDROCARBONS AND NITROGEN OXIDES

There are substantial uncertainties in even qualitatively predicting how simultaneously reducing  $\text{SO}_2$ ,  $\text{NO}_x$ , and RHC emissions from given sources would affect acid deposition at a distant receptor site. Altering the concentrations of RHC and  $\text{NO}_x$  in an urban plume that interacts with  $\text{SO}_2$  from a powerplant plume may well alter the amount of acid deposition at a particular downwind location, but the meteorological and chemical factors involved are so complex that no reliable quantitative estimates can be made at the present time.

For example, changing RHC and  $\text{NO}_x$  emissions *without* changing  $\text{SO}_2$  emissions could affect the deposition of sulfuric acid at a given location by changing the concentrations of available oxidants needed for converting  $\text{SO}_2$  to sulfate. Further effects could arise if alterations in RHC and  $\text{NO}_x$  concentrations affected the pH

\* Peak ozone levels decrease because at higher  $\text{NO}_x$  concentrations, a larger portion of the available free radicals react with  $\text{NO}_2$  to form nitric acid, and become unavailable for the chain reactions leading to ozone formation.

Figure C-3.—Typical Ozone Concentrations Formed From RHC-NO<sub>x</sub> Mixtures

Oxidant/O<sub>3</sub> isopleths derived from combined use of smog chamber and photochemical modeling techniques.

SOURCE: *Air Quality Criteria for Ozone and Other Photochemical Oxidants*, U.S. Environmental Protection Agency, Research Triangle Park, EPA-600/8-78-004, April 1979

of existing cloud droplets, e.g., through the formation and dissolution of nitric acid in the cloud. For the aqueous phase oxidation of SO<sub>2</sub>, in which the major processes are pH-dependent, changes in cloud acidity levels could change the rate of sulfuric acid formation.

If the net result of changed RHC and NO<sub>x</sub> emissions is faster conversion of SO<sub>2</sub>, sulfuric acid will form and be deposited closer to sources. If the concentrations of RHC and NO<sub>x</sub> are altered such that less oxidizing material is initially available, sulfuric acid is deposited further from emission sources. Therefore, alterations in the rate of oxidation could change the amount of acid deposition to a specific location, but not the *total* amount of sulfur deposited (e. g., the sum of SO<sub>2</sub> and sulfates) over the entire downwind area. The sulfur emitted will eventually return to the surface at some point. If NO<sub>x</sub> and RHC emissions are decreased such that their ratio remains constant while SO<sub>2</sub> emissions are decreased, total deposition of both sulfuric and nitric acid will decrease and total ozone production will decrease.

#### INCREASE NITROGEN OXIDES, HOLDING HYDROCARBON AND SULFUR DIOXIDE EMISSIONS CONSTANT

This scenario is likely to occur without major changes in current air pollution control regulations. Future increases in NO<sub>x</sub> levels are projected (primarily from utilities and the industrial sector), while SO<sub>2</sub> emissions are projected to remain fairly constant or increase slightly over the next 20 years (see app. A). Chamber studies show that increasing NO<sub>x</sub> from very low levels, holding RHC levels constant, causes *peak* ozone concentrations to increase. As NO<sub>x</sub> levels are further increased, ozone concentrations reach a maximum and then decrease with further increases in NO<sub>x</sub> (see fig. C-3).

The concentrations of other nitrogenous pollutants such as nitrous acid also generally increase with increased NO<sub>x</sub>. Increasing NO<sub>x</sub> emissions while SO<sub>2</sub> emissions are held constant will augment local acid deposition due to nitric acid. Since emitted NO<sub>x</sub> are oxidized more readily than SO<sub>2</sub> in the gas phase, increas-

ing  $\text{NO}_x$  concentrations may expand the geographical area over which sulfur deposition occurs.

### Source-Receptor Relationships

A major goal of atmospheric science is to predict for a given pollution source, the dose of pollutants at a specific location downwind, and how changes in that source's emissions would change the pollutant burden at the receptor site. Source-receptor relationships are determined by the location and nature of the primary pollutant emissions (e. g.,  $\text{SO}_2$ ,  $\text{NO}_x$ , RHC) and by associated meteorological, chemical, and physical processes that occur as the pollutants travel from the source to the receptor. Current long-range transport models that incorporate sophisticated meteorology (i. e., those used in the Canadian-American Work Group effort under the Memorandum of Intent) attempt to simulate chemical conversions of  $\text{SO}_2$  to sulfate by assuming that the complex set of chemical processes will balance out over time and distance to approximate a constant average rate of transformation. This simplifying assumption makes regional-scale calculations tractable; crude source regions and receptor regions can be identified for sulfur compounds. Because they are linear models, reducing emissions in source regions results in a proportional reduction of deposition in receptor regions. These models appear to characterize the *current* situation for wet deposition fairly well. They use actual emissions as input, and can reproduce observed levels of regional wet sulfate deposited within a factor of 2.

The first attempt to incorporate multiple chemical reactions involving  $\text{NO}_x$ ,  $\text{SO}_2$ , and RHC into a long-range transport model—called the Rohde model—contains 19 chemical equations but virtually no meteorology. Three of the equations concern sulfuric or nitric acid production. Sixteen of the equations describe the gas phase photochemistry associated with the RHC/ $\text{NO}_x$  systems, ozone, hydroxyl radical, and hydrogen peroxide—all compounds involved in actually forming the acid. All of the aqueous and heterogeneous phase reactions are combined into one simplified equation.

This model assumes that dry deposition decreases in proportion to emissions. However, it predicts that reducing emissions might cause wet deposition to decline less than proportionally -e.g., a response 60 percent as great.\*

Recently, a Committee of the National Academy of Sciences altered the chemistry in the Rohde model to incorporate new laboratory results. It found that the new assumptions greatly reduced the nonlinearity in the rela-

tionship between ambient  $\text{SO}_2$  concentrations and ambient sulfate concentrations. Using currently available data, the NAS report concludes that, "there is no evidence for strong nonlinearity in the relationships between long-term average emissions and deposition.

Very specific source-receptor relationships cannot be defined unless the behavior of all other pollutants is known. Given the complexity of the atmospheric chemistry alone, as well as the need to develop detailed emission inventories (especially for  $\text{NO}_x$  and RHC), and inherent meteorological variability, it is unlikely that a definitive model integrating  $\text{SO}_2$ ,  $\text{NO}_2$ , and RHC will be developed in the next decade or two. Decisions to control or not to control precursor emissions over this time period will have to be made without the benefit of such precise information.

### Selected References

The following list of references includes recent review articles and several very recent key papers that may not be included in the review articles cited.

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\*See appendix C.2, "Source-Receptor Relationships," for further explanation. Note that OTA specified the model with conservative assumptions; subsequent runs using different background concentrations for pollutants over time show that wet deposition may respond more directly to emissions reductions.

<sup>1</sup>Acid Deposition, *Atmospheric Processes in Eastern North America*, Committee on Atmospheric Transport and Chemical Transformation in Acid Precipitation, National Research Council (Washington, D. C.: National Academy Press, 1983).



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## C.2 SOURCE-RECEPTOR RELATIONSHIPS

### Introduction

Broad regions of North America receive acidic deposition both in wet form—acid rain—and as dry deposition of acidic substances. Because acidifying substances reach the Earth from the atmosphere both through removal by rainfall and as directly deposited gases and particles from the air, the term acid rain is misleading. The acidity of rainfall per se is generally considered less significant than the quantity of ‘acid-producing’ substances added to the environment—onto soils, vegetation, and materials, and, after passing through watersheds, into lakes and streams. The term acid-producing substances as used in this report refers to sulfur oxides ( $\text{SO}_x$ ) and nitrogen oxides ( $\text{NO}_x$ ) and other substances that have the *potential* for producing acidity, although they may not be deposited in an acid form. For example, sulfate can be deposited in a neutral form—ammonium sulfate—but end up as sulfuric acid by the time it reaches a lake or stream if the ammonium is used by plants in the watershed.

The chain of physical and biological processes from emissions of pollutants to eventual deposition of acid-producing substances in the environment is complex and not fully understood. However, several lines of evidence can be combined to express the likely relationship between emissions and deposition.

Acid deposition results from both local and distant sources of  $\text{SO}_x$  and  $\text{NO}_x$ . Current scientific understanding suggests that reducing sulfur dioxide ( $\text{SO}_2$ ) emissions throughout the Eastern 31-State region would reduce the deposition of acid-producing substances; but that this will occur in the areas *sensitive* to acid deposition cannot be stated with certainty. For the *Eastern United States*, no other control strategy offers greater potential for reducing acid deposition than reducing  $\text{SO}_2$  emissions. While curbing other pollutant emissions could be considered simultaneously, and might ultimately be necessary to achieve a desired level of deposition reduction, most scientists would focus initial attempts to control acid deposition on  $\text{SO}_2$ .

By considering preliminary information drawn from several alternative approaches, one can piece together a *plausible* relationship between pollutant emissions and deposition of acid-producing substances. This appendix addresses four key issues. The discussion begins with regional-scale deposition of acidifying substances, and then identifies the major constituents of deposition and the relative magnitude of current sources. Model-based estimates of how pollutant emissions from parts of the Eastern United States affect deposition in other areas

are then discussed. Two complementary modeling approaches are used to estimate the magnitude of potential reductions in acid-producing substances reaching the environment due to reductions in  $\text{SO}_2$  emissions. Finally, the analysis addresses the question of whether these potential reductions in acid-producing substances might be enough to meet ‘target’ deposition rates to protect sensitive resources.

### Current Deposition of Acid-Producing Substances

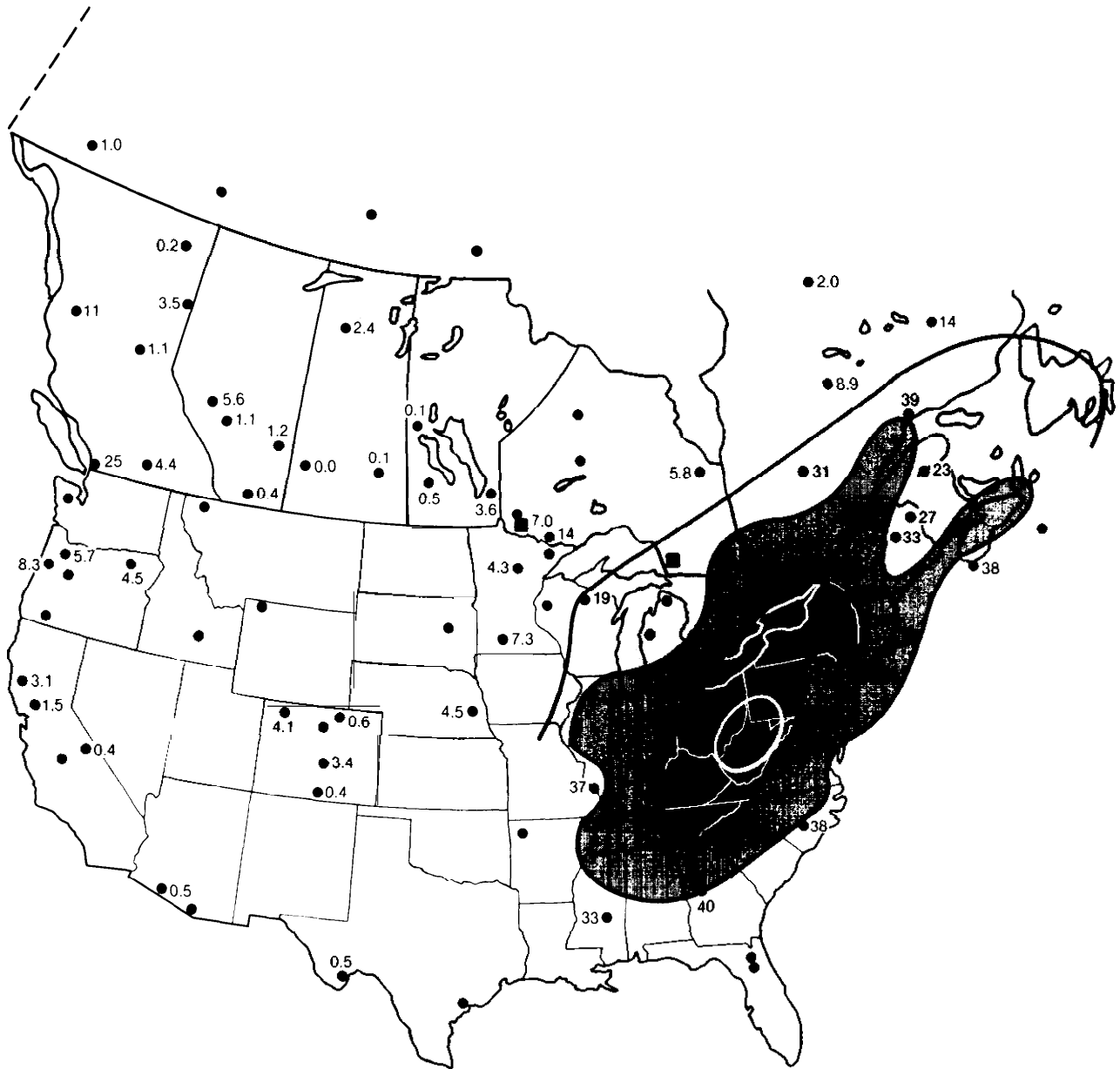
The best information on patterns of acidic deposition—both its chemical composition and spatial distribution over affected parts of North America—comes from monitoring networks collecting rainfall samples. Though wet deposition may account for only about half the deposited  $\text{SO}_x$  and  $\text{NO}_x$ , “dry-deposited gaseous and particulate pollutants are not monitored extensively enough to determine their precise distribution. This discussion focuses primarily on acid-producing substances deposited through precipitation.

The balance of chemical species originating from such natural sources as seaspray, windblown soil, and carbon dioxide in the air, and such manmade sources as  $\text{SO}_x$  and  $\text{NO}_x$  pollutants, determine the acidity of rainfall. Rainfall acidity can be decreased either by removing acid-producing substances, or by adding acid-neutralizing substances. The major acid-producing substances in rainfall in the Eastern United States are  $\text{SO}_x$  and  $\text{NO}_x$  from both natural and manmade sources. However, the presence of these substances in rainfall does not necessarily indicate acidity since they can be counterbalanced by such airborne acid-neutralizing substances as calcium and magnesium from soil and ammonium from natural sources and fertilizers.

Patterns of acidity, sulfate, and nitrate deposition in rainfall over Eastern North America are mapped in figures C-4 through C-6. Figure C-4 presents the deposition of hydrogen ions—the substance actually measured to determine acidity. More than 40 milliequivalents per square meter per year ( $\text{meq}/\text{m}^2/\text{yr}$ ) of hydrogen ions are

● Most long-range transport air pollution models estimate that dry deposition is about equal to wet deposition when averaged over the Eastern United States. The ratio of dry to wet deposition varies with distance away from sources of pollution, with ratios on the order of 8 to 12 in areas with high  $\text{SO}_2$  concentrations, to about 0.2 to 0.4 in the Adirondack Mountains (D. Fowler, “Removal of Sulphur and Nitrogen Compounds From the Atmosphere, Ecological Impact of Acid Precipitation, SNSF project, October 1980. A.H. Johannes, et al., “Relationships Between Wet Deposition, Dry Deposition and Through-fall Chemistry,” Air Pollution Control Association Annual Meeting, New Orleans, June 1982.

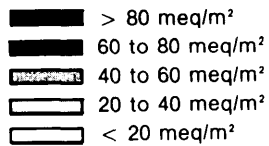
Figure C-4.—Hydrogen Ion in Precipitation, 1980 (annual deposition in milliequivalents per square meter)



Legend:

Data compiled from Canadian and American monitoring networks.

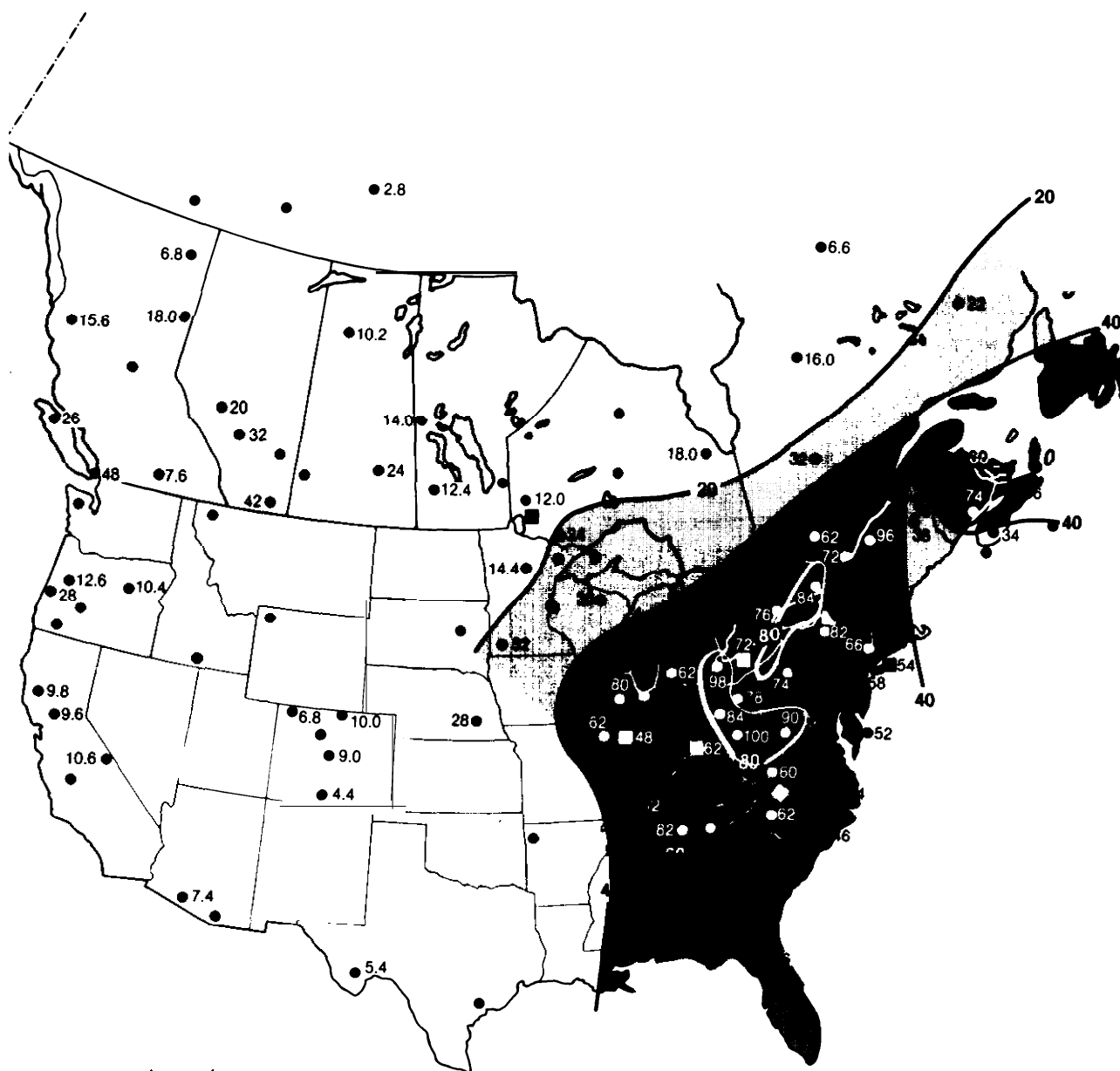
Canada: CANSAP  
 APN  
 OME  
 United States: NADP  
 MAP3S



1 millequivalent/meter<sup>2</sup> =  
 0.01 kilograms/hectare

SOURCE: Impact Assessment, Work Group 1, United States-Canada Memorandum of Intent on Transboundary Air Pollution, final report, January 1983

**Figure C-5.—Sulfate in Precipitation, 1980 (annual deposition in milliequivalents per square meter)**



**Legend:**

Data compiled from Canadian and American monitoring networks.

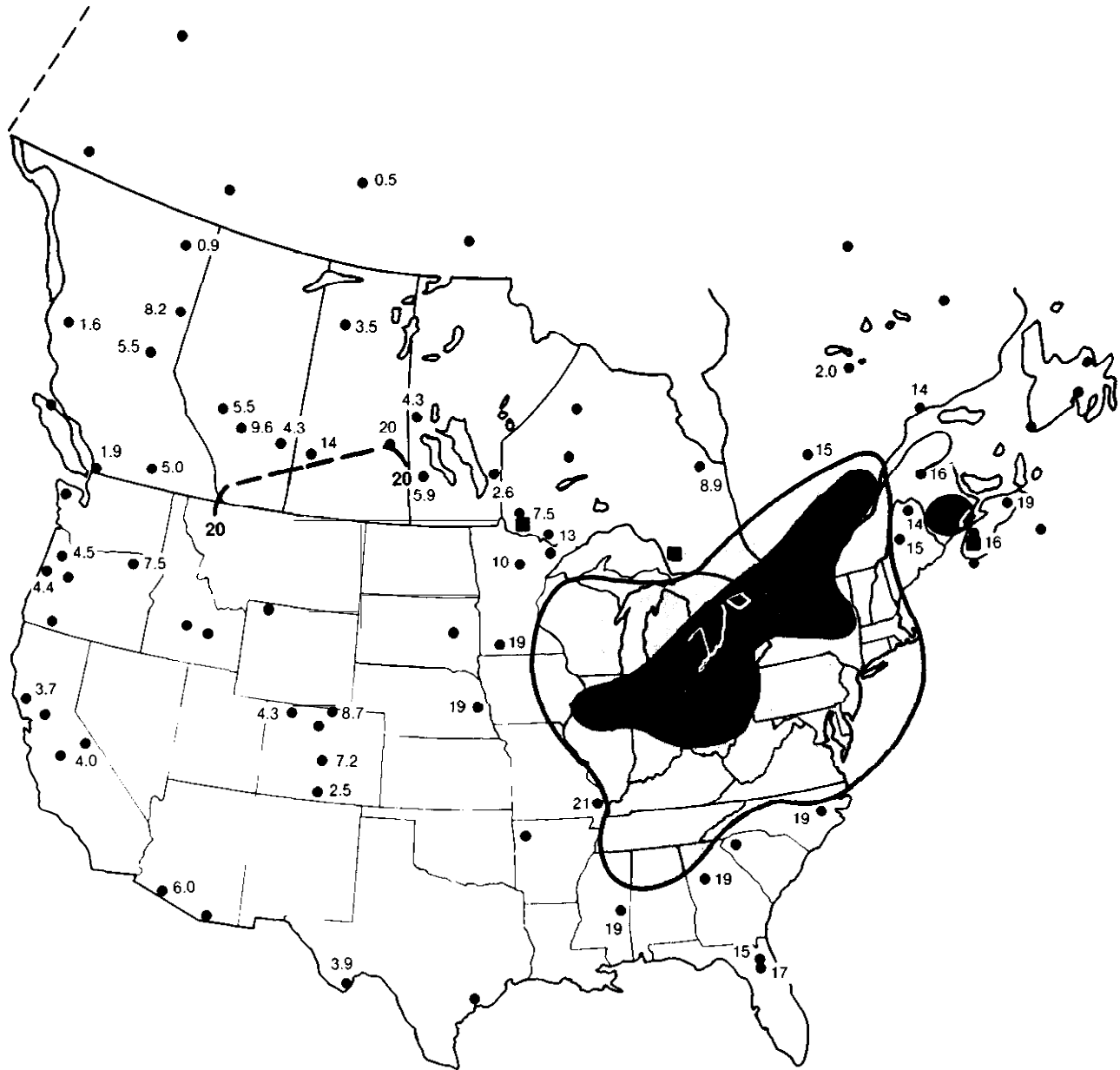
- |          |                |
|----------|----------------|
| Canada:  | United States: |
| ● CANSAP | ● NADP         |
| ■ APN    | ■ MAP3S        |
| ◆ OME    |                |

- > 80 meq/m<sup>2</sup>
- 60 to 80 meq/m<sup>2</sup>
- 40 to 60 meq/m<sup>2</sup>
- ▬ 20 to 40 meq/m<sup>2</sup>
- ▬ < 20 meq/m<sup>2</sup>

1 millequivalent/meter<sup>2</sup> = 0.48 kilograms/hectare

SOURCE: Impact Assessment, Work Group 1, United States-Canada Memorandum of Intent on Transboundary Air Pollution, final report, January 1983.

Figure C6.—Nitrate in Precipitation During 1980  
(weighted by precipitation-milliequivalents per square meter)



Legend:

Data compiled from Canadian and American monitoring networks.

- |          |                |
|----------|----------------|
| Canada:  | United States: |
| ● CANSAP | ● NADP         |
| □ APN    | □ MAP3S        |
| ◇ OME    |                |

- > 80 meq/m<sup>2</sup>
- 60 to 80 meq/m<sup>2</sup>
- 40 to 60 meq/m<sup>2</sup>
- 20 to 40 meq/m<sup>2</sup>
- < 20 meq/m<sup>2</sup>

1 millequivalent/meter<sup>2</sup> =  
0.62 kilograms/hectare

SOURCE Impact Assessment, Work Group 1, United States-Canada Memorandum of Intent on Transboundary Air Pollution, final report, January 1983

deposited over broad regions of the Eastern United States, in contrast to deposition rates under 10 meq/m<sup>2</sup>/yr throughout much of the West. The highest deposition rates exceed 80 meq/m<sup>2</sup>/yr, and are centered around eastern Ohio, western Pennsylvania, and northern West Virginia.

Of the major acid-producing substances in rainfall, only two originate in substantial amounts from manmade sources in the Eastern United States: sulfates and nitrates. The patterns of deposited acidity and deposited sulfate (fig. C-5), are quite similar. A larger part of the Eastern United States receives deposition in excess of 40 meq/m<sup>2</sup>/yr. The highest deposition rates again exceed 80 meq/m<sup>2</sup>/yr, centered in about the same region. Nitrate deposition patterns are similar (figs. C-5 and C-6), but deposition rates are about one-half those for sulfur. More than 20 meq/m<sup>2</sup>/yr of nitrates are deposited over broad regions of the Eastern United States, with peak deposition exceeding 40 meq/m<sup>2</sup>/yr. About two-thirds of the total sulfate and nitrate deposited in the Eastern United States is sulfate.

By comparing figures C-4 through C-6, one can see that the deposition of *acid-producing substances* (sulfate and nitrate) exceeds the deposition of *acidity* by about 25 to 50 percent; this portion of the sulfate and nitrate is neutralized by such other constituents as calcium and ammonium.

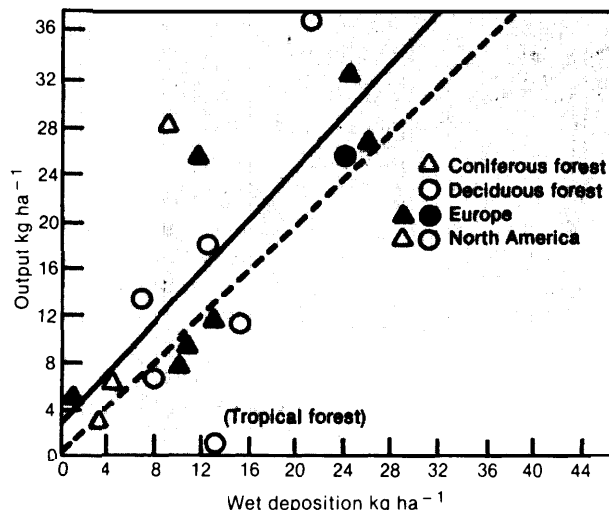
Reducing sulfate deposition might be the most likely way to begin reducing the deposition of acidity, given that about twice as much sulfate as nitrate is present in rainfall. Similar patterns are assumed to occur for dry deposition of acid-producing substances, but far fewer observations are available to substantiate this.

The most significant indicator for assessing alterations in lake- and stream-water quality is the acidity of water after it flows through a surrounding watershed—i.e., the total amount of acid-producing substances that eventually reach and travel through aquatic environments. Figure C-7 illustrates the relationship between the quantity of acid-producing substances in rainfall and the quantity of acid-producing substances in water flowing out of a watershed. Figure C-7A shows that the amount of sulfate leaving a watershed is about equal to, and in many cases greater than, the amount entering a watershed in rainfall, when averaged over a period of one or several years. That more sulfur leaves a watershed than enters from rainfall is probably due to the amounts that enter as dry-deposited gases and particles.

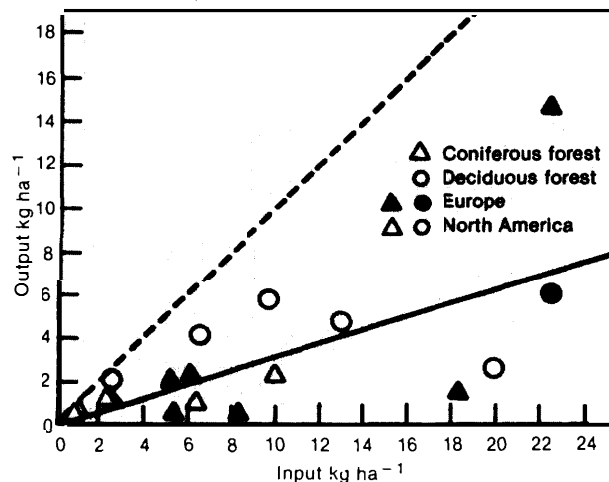
For nitrogen-containing substances such as nitrate and ammonium (as shown in fig. C-7 B), the picture is different. Only about one-third of the nitrogen entering a watershed from rainfall leaves the watershed. If the nitrogen input from dry deposition is included, the ratio of nitrogen leaving to nitrogen entering a watershed becomes even lower. As discussed in the forest

**Figure C-7.—Sulfur and Nitrogen: Quantities in Precipitation Versus Quantities in Outflow From Watersheds**

A: Annual input of SO<sub>4</sub><sup>2-</sup> - S by precipitation versus the leaching losses from forest watersheds and lysimeters with forest soils. Solid line is the regression line, dotted line is the 1:1 line.



B: Annual input of (NH<sub>4</sub><sup>+</sup> + NO<sub>3</sub><sup>-</sup>) - N by precipitation versus the leaching losses from forest watersheds and lysimeters with forest soils. Solid line is the regression line, dotted line is the 1:1 line. Data from studies in Europe and North America.



NOTE: Solid line is the regression line, dotted line is the 1:1 line. Data from studies in Europe and North America.

SOURCE: G. Abrahamson, "Acid Precipitation, Plant Nutrients, and Forest Growth" (1980). Ecological Impact of Acid Precipitation, Proceedings of an International Conference.

resources section, these results might be expected—while both nitrogen and sulfur are essential nutrients for forest growth, most eastern forests require far greater inputs of nitrogen than sulfur.

In summary, about twice as much sulfate is generally present in rainfall as nitrate. When considering the

acid-producing substances that travel *through the environment* (and effects such as altered lake- and stream-water quality), sulfate may predominate even more. This can vary from region to region, and certainly varies for short periods. For example, the nitrate component of spring snowmelt can be as great as or even greater than sulfate levels in some watersheds. However, over broad regions on an annual time scale, sulfates comprise a much larger share of acid-producing substances than nitrates.

#### Manmade and Natural Source Contributions to Acidic Deposition

Relatively low background levels of acidic deposition are thought to originate from natural sources (and possibly global-scale transported air pollutants), when averaged over large regions (e. g., the Eastern United States or the North American Continent). This “natural background” deposition of wet sulfur in North America (excluding Mexico) has been estimated to be about 4 to 10 meq/m<sup>2</sup>.<sup>4</sup> This is about 20 to 40 percent of the *average* sulfur deposition from precipitation over all of North America. Natural sources contribute relatively smaller proportions in areas of highest deposition. Averaged over Eastern North America (east of the Mississippi and south of James Bay in Canada), natural background sources might contribute about 12 to 25 percent of the total. Another group has estimated natural sources of sulfur to be about 5 to 10 percent of manmade sources in this same region.<sup>5</sup> Over parts of the Eastern United States, where wet sulfur deposition exceeds 50 meq/m<sup>2</sup> and is as high as 80 meq/m<sup>2</sup> in some areas, the natural background contributes even less.

This natural background comes from several different sources: Husar estimates that 6 percent of the wet sulfur deposition in North America originates from seaspray. The Electric Power Research Institute (EPRI) has estimated that about 5 percent of SO<sub>2</sub> emissions originate from natural, biological sources.<sup>6</sup> In addition, geologic sources such as volcanoes are thought to contribute to natural background.

When averaged over the North American Continent, the acid-producing *potential* of manmade SO<sub>2</sub> emissions far exceeds total wet sulfur deposition. The over 30 million tons of SO<sub>2</sub> emitted by manmade sources in North America is 2.5 times total wet sulfur deposition and 6 to 10 times the natural background. The portion

of manmade sulfur that is not deposited in rainfall is either deposited dry or exported off the continent.

Natural background is estimated to contribute similar proportions of total wet nitrate deposition—about 20 to 40 percent when averaged over the North American Continent, and about 10 to 25 percent when averaged over Eastern North America.

#### The Relationship Between Current Emissions and Deposition

One of the major controversies in the acid rain debate is the effect that pollutant emissions from any source (or group of sources) will have on ambient air quality and pollutant deposition at some other location. The relationship between emissions and deposition is determined by a complex chain of chemical and physical processes that occur as primary pollutants (e. g., SO<sub>2</sub>) are emitted, transformed into secondary pollutants (e. g., sulfates), transported, and finally deposited.

Computer models, called transport models, are used to mathematically *simulate* the transformation, transport, and deposition processes. Models describing long-range transport of SO<sub>2</sub> have been available for several years; preliminary models of NO<sub>x</sub> transport are just now being developed. Transport models are the only practical procedure available to estimate the relationship between areas of origin and areas of deposition for long-range transport pollutants, unless newly developed tracer techniques prove reliable. Large-scale regional transport cannot now be measured directly for the large number of sources of emissions and deposition regions of interest, and under the variety of meteorological conditions needed to perform the analysis.

The major long-range transport (LRT) models describing SO<sub>x</sub> transport incorporate six atmospheric processes:

1. release of emissions,
2. horizontal transport and dispersion,
3. vertical mixing of pollutants in the atmosphere,
4. chemical transformation,
5. dry deposition, and
6. wet deposition.

The models themselves are composed of submodels which simulate chemical and physical processes within these broader categories. The main data requirements of LRT models are: emissions inventories of pollutants, meteorological data, ground cover data, and a host of values (parameters) representing chemical reaction rates and other physical processes.

Eight LRT models were evaluated under the U. S.-Canada Memorandum of Intent (MOI) on Transboundary Air Pollution;<sup>7</sup> results indicate that the models

<sup>4</sup>R. B. Husar and J. M. Holloway, “Sulfur and Nitrogen Over North America, presented at 1982 Stockholm Conference on Acidification of the Environment

<sup>5</sup>*Atmospheric Sciences and Analysis, Work Group 2, United States-Canada Memorandum of Intent on Transboundary Air Pollution, Final Report, November 1982*

<sup>6</sup>Electric Power Research Institute, “Biogenic Sulfur Emissions in the SURE Region’ EA-1516, 1980

<sup>7</sup>*Atmospheric Sciences and Analysis, op cit,*

appear to reproduce large-scale patterns of observed wet sulfur deposition. However, the size and quality of the data base precludes a complete evaluation of LRT models at this time. \*

This section presents modeling results to provide a *plausible* description of the current relationship between sources and deposition of SO<sub>x</sub> in the Eastern United States. This is only one of the acid-producing substances currently deposited; however, as discussed earlier, sulfate is currently the major acid-producing substance in precipitation.

The model used in this analysis is the Advanced Statistical Trajectory Regional Air Pollution Model (ASTRAP) developed by Argonne National Laboratory under DOE and EPA funding. The ASTRAP model includes several components of the source-receptor relationship not included in the other models. \* \*

OTA has used another model that incorporates more realistic atmospheric chemistry and considers the effects of co-pollutants such as NO<sub>x</sub> and hydrocarbons (but at the expense of sophisticated meteorology), to assess how the simplifying chemical assumptions used in ASTRAP might affect its resulting projections. This comparison indicates that the ASTRAP model might adequately represent dry deposition of sulfur, but that variations in ambient concentrations of other pollutants might significantly affect wet sulfur deposition. (These results are discussed in the preceding subsection, "Atmospheric Chemistry.")

Because of its sophisticated treatment of the regional patterns of emissions and meteorology, the ASTRAP model can be used to investigate the *current* relationship between regions of SO<sub>2</sub> emissions and regions of sulfur deposition. The model is best used in a relative sense—e.g., estimating the proportion of deposition one region contributes to another—rather than for projecting the magnitude of deposition quantitatively: Again the following discussion of sulfur deposition describes the general pattern of current relationships, and must not be interpreted as making quantitative predictions.

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"Even if the models were 'perfect,' one would expect the model simulations to deviate from the observations since the latter are influenced by both the factors treated in the model and the factors that are not treated (e. g., small-scale precipitation variations). In addition, part of the difference between model simulations and observations is due to the inherent variability of the real world. The model is of necessity designed to simulate an average of the observations while the monitoring data base at this time is insufficient to calculate a representative average (i. e., the average over a number of years with similar emissions, meteorological conditions, etc.).

- "The ASTRAP model includes: the ability to account for seasonal differences (the ASTRAP model simulates January and July average conditions); release height of emissions; the use of detailed meteorological data; consideration of vertical atmospheric processes in addition to horizontal movement; wet deposition rates that vary with rainfall intensity; and dry deposition rates that vary temporally and spatially. The other models evaluated include some of these components, but ASTRAP is the only model that includes all of them.

Acid deposition has often been characterized as 'acid in Adirondack lakes from powerplants in Ohio. The ASTRAP model can be used to show that such statements are overly simplistic: pollutants do travel from one region to another, but in all directions, not just west to east. In addition, while pollutants can travel long distances, emissions *within* a region contribute a large share to total deposition in that region.

To illustrate these points, figure C-8 divides eastern North America into four regions. The intersection of the regions has been chosen to correspond to the area of peak wet sulfur deposition in 1980. Figure C-8 also displays the percentage of SO<sub>2</sub> emitted in each region, and the percentage of total sulfur deposited (as simulated by ASTRAP) in each of the regions. Sulfur dioxide emissions are roughly comparable in the northeastern region (I), southeastern region (II), and southwestern region (III). Emissions in the northwestern region (IV) are over twice the amount of any of the other regions. Deposition is lowest in the southern regions (II and III) and highest in the northern regions (I and IV).

Figures C-9 through C-12 show model-based estimates of: 1) the percentage of each region's deposition originating from *within* its borders, and from each of the *other* regions; and 2) for deposition originating *outside* a region, the percentage of deposition traveling less than 500 km, 500 to 1,000 km, 1,000 to 1,500 km, and greater than 1,500 km.

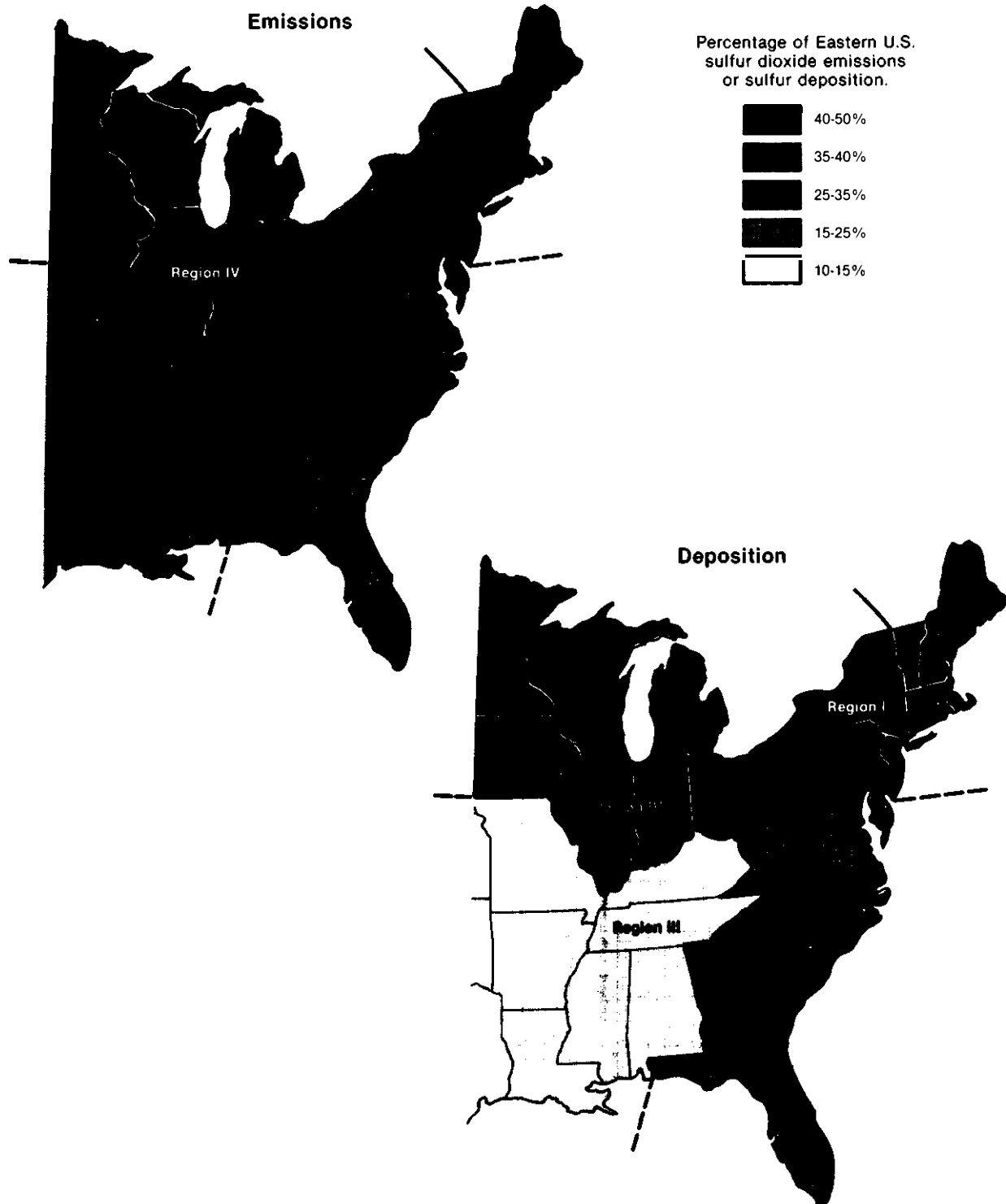
For example, figure C-9 illustrates these relationships in region I (the northeastern region). The pie chart in the upper right projects that approximately 80 percent of the deposition comes from emissions in two regions— from within its own borders and from the northwestern region in about equal amounts. The bar graphs placed in regions II, III, and IV illustrate each region contribution to deposition in the northeastern region, according to its distance from the sources of emissions. For example, the bar graph in the lower right shows model estimates that 40 percent of the deposition coming from the southeastern region travels less than 500 km to its eventual area of deposition in the northeast; another 40 percent travels between 500 to 1,000 km, and the remainder travels over 1,000 km.

Figures C-9 through C-12 demonstrate the following general observations:

1. All regions contribute to deposition in all other regions.
2. At the spatial scale used in this analysis, each region generates as much or more of its own deposition as any other single region contributes to it.
3. Substantial quantities of deposition originate from sources over 500 km away.
4. Pollutants are transported further from west to east and south to north than from east to west or north to south.

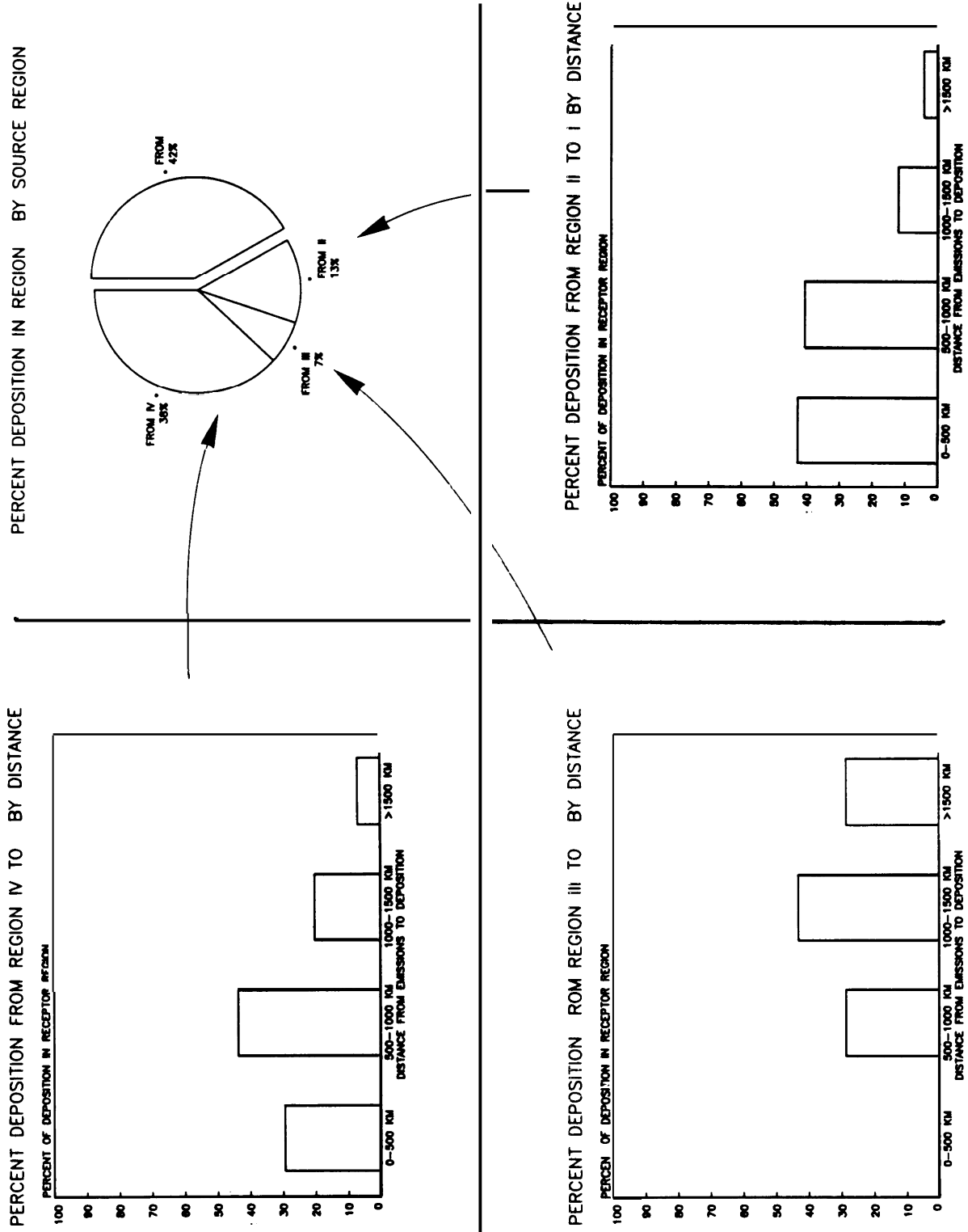


Figure C-8.—1979 Sulfur Dioxide Emissions and Estimated Sulfur Deposition—Percent Contributed and Received in Four Subregions Covering the Eastern Half of the United States



SOURCE: J Shannon, personal communication, Argonne National Laboratory and E. H. Pechan & Associates, Inc., 1982

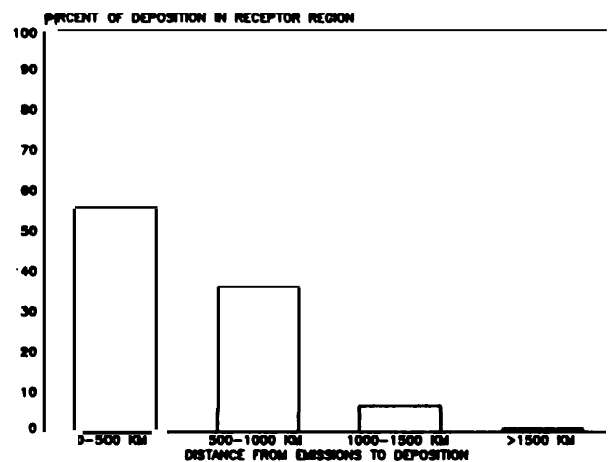
Figure C-9.—Deposition in Region



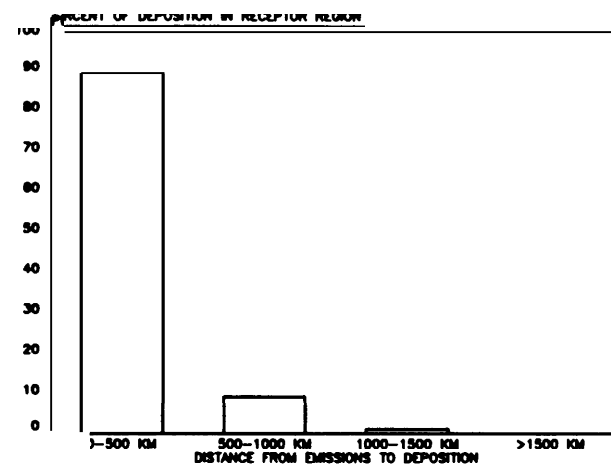
SOURCE: J. Shannon, personal communication, Argonne National Laboratory and E. H. Pechan & Associates, Inc., 1982.

Figure C-10.—Deposition in Region II

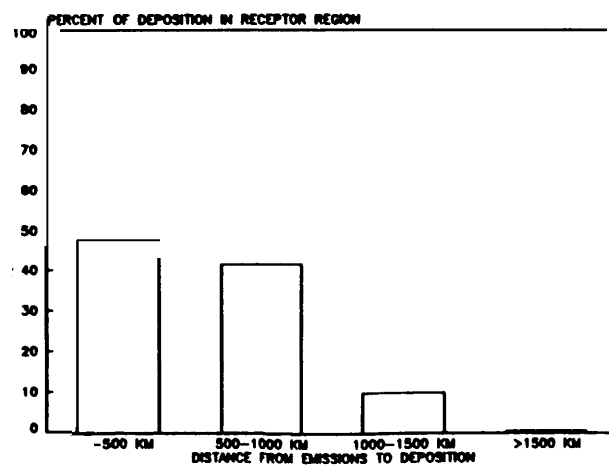
PERCENT DEPOSITION FROM REGION IV TO II BY DISTANCE



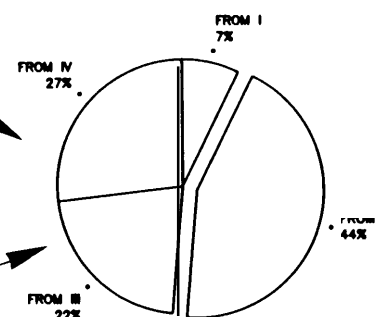
PERCENT DEPOSITION FROM REGION I TO II BY DISTANCE



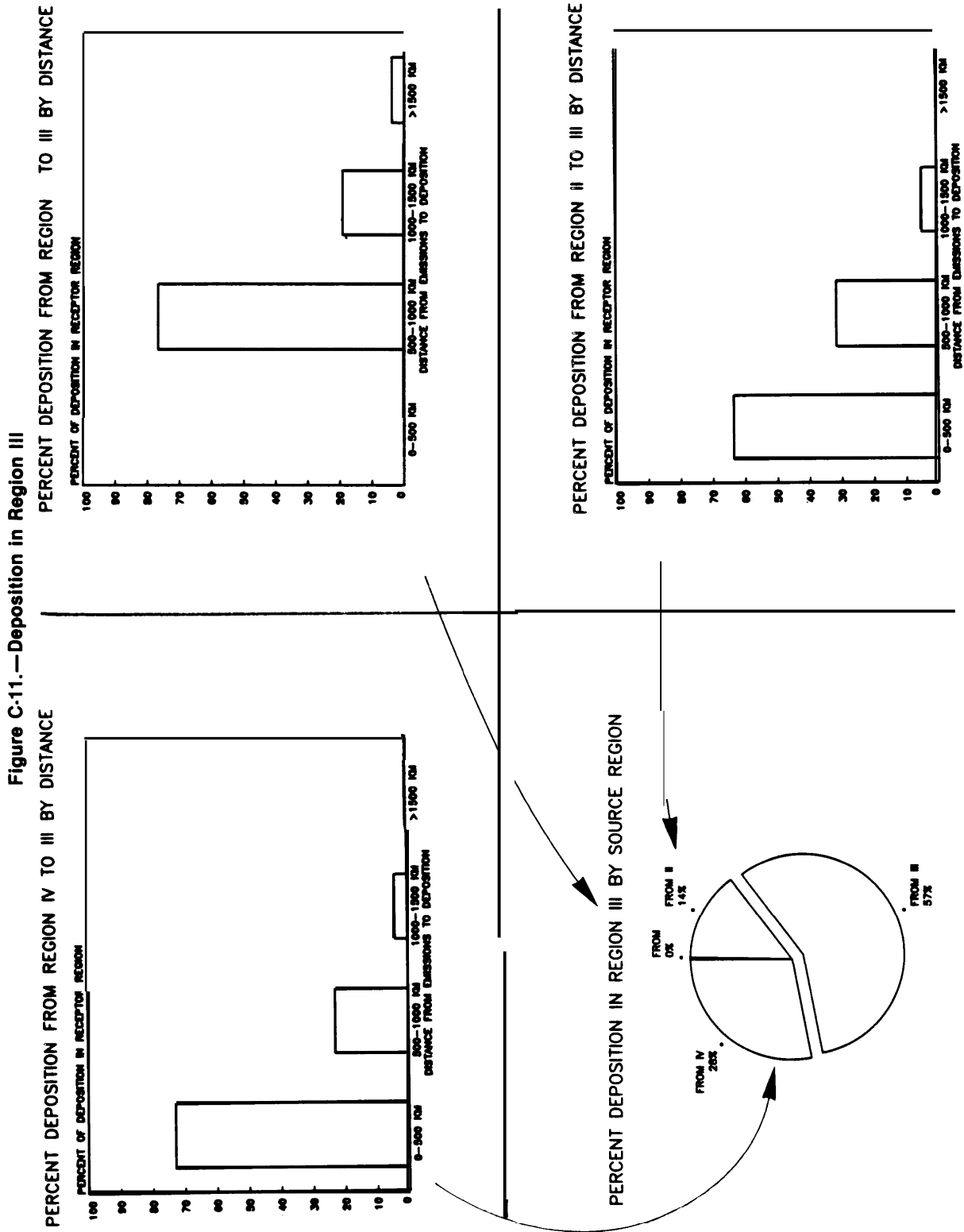
PERCENT DEPOSITION FROM REGION III TO II BY DISTANCE



PERCENT DEPOSITION IN REGION II BY SOURCE REGION

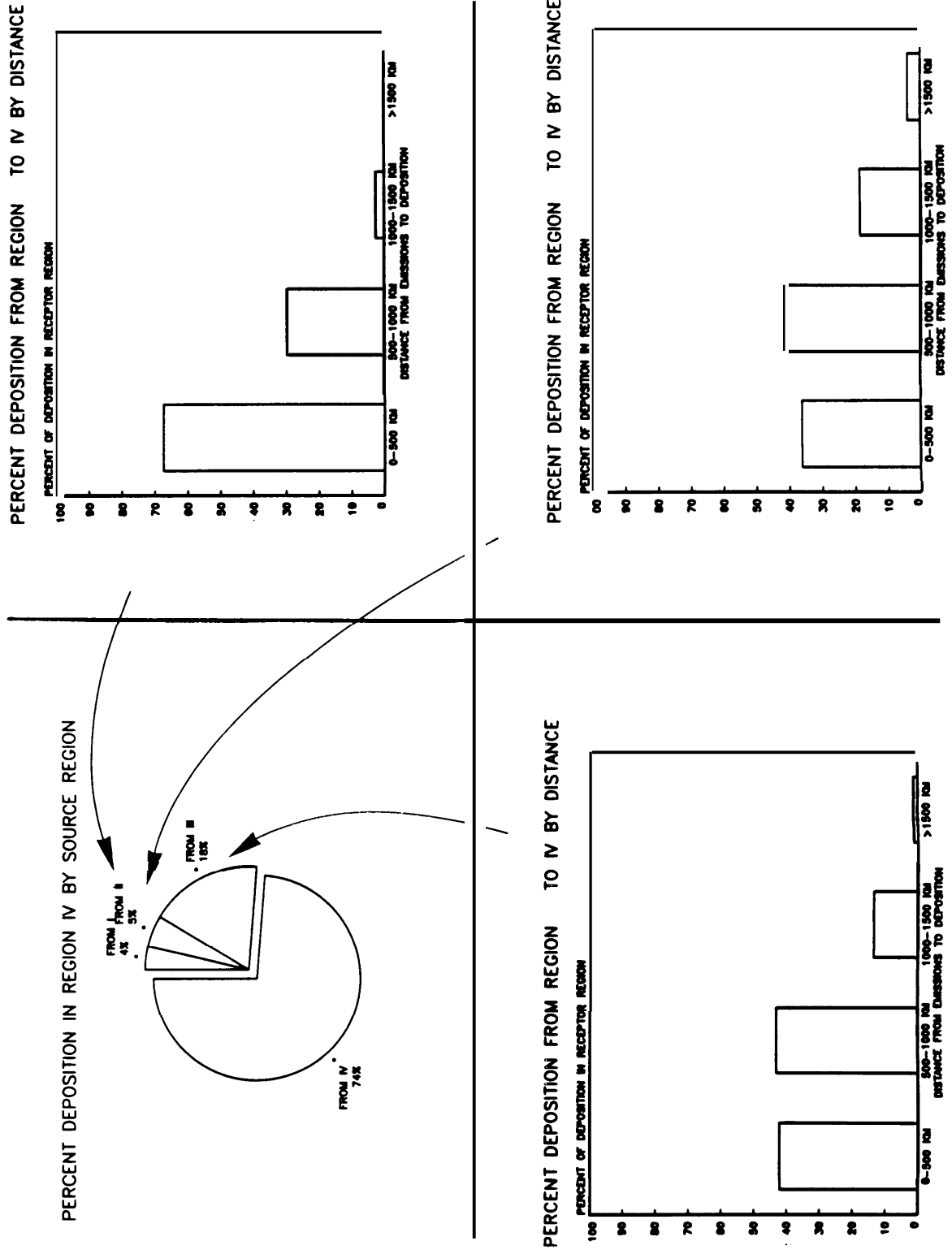


SOURCE: J. Shannon, personal communication, Argonne National Laboratory and E. H. Pechan & Associates, Inc., 1982



SOURCE: J. personal communication, Argonne Laboratory and H. Pechan & Associates, Inc., 1982

Figure C-12.—Deposition in Region IV



SOURCE: J. personal communication, Argonne National Laboratory and E. Pechan & Associates, Inc., 1982.

## The Effectiveness of Emissions Reductions for Achieving Deposition Reductions

**Assumptions concerning the physical and chemical processes involved in transforming SO<sub>2</sub> to sulfate are inherent in the use of regional-scale models such as ASTRAP.** Linear regional-scale models assume that sulfate production is proportional to the concentration of SO<sub>2</sub>. The source-receptor relationships described by these models can simulate expected changes in deposition only to the extent that deposition would actually change in linear proportion to changes in emissions.

While the preceding source-receptor relationships may provide reasonable estimates of inter- and intra-region transport, they may not be reliable for developing control strategies unless chemical transformation and deposition processes can be shown to behave in a linear manner. This section first discusses linear model projections of the effects of different emissions scenarios and then considers how the addition of more realistic atmospheric chemistry might alter the results.

OTA used the ASTRAP model to simulate atmospheric concentrations and surface deposition of sulfur pollutants for three alternative levels of SO<sub>2</sub> emissions. \* These include: current emissions levels in the United States and Canada; a representative 8-million-ton-per-year emissions reduction; and a representative 10-million-ton-per-year emissions reduction. \* \*

The model was used to simulate deposition levels during January and July, to investigate the effects of both winter and summer conditions. Figure C-13 displays model projections of how extensively both wet and dry sulfur deposition would be reduced by a representative 10-million-ton reduction in SO<sub>2</sub> emissions. The reductions shown apply only to emissions originating in the United States (i.e., deposition from emissions in Canada are not considered). Similar projections for an 8-million-ton reduction are shown in figure C-14. The pattern of deposition reductions is similar for both scenarios. Deposition is reduced by the greatest percentage in the Midwest—the region with both the highest current emissions and the greatest reduction requirements under both scenarios. Proportional reductions in deposition decline with distance away from this region. Because the ASTRAP model assumes a linear relationship between emissions and deposition, both scenarios show

\* Personal communication, J. Shannon, Argonne National Laboratory, December 1981.

\* Senate bills S. 1706 and S. 1709 (97th Congress) were two of the earliest legislative proposals to control acid deposition. S. 1706 (introduced by Senator Mitchell of Maine) and S. 1709 (introduced by Senator Moynihan of New York) were both referred to the Senate Committee on Environment and Public Works October 1981. The State-level reductions specified by these bills were used as the basis for the 10- and 8-million-ton reduction scenarios, respectively. These scenarios assume that reductions in emissions from each point source within a State are proportional to the reduction assigned that State in the OTA analysis of these bills.

that *total* deposition is reduced almost in proportion to reductions in total emissions when averaged over the entire Eastern United States.

Assuming a linear relationship—i.e., that a specified percentage reduction in emissions will lead to the same percentage reduction in deposition—is a simplification adopted for computational advantages. No one would argue that this simplified relationship realistically represents the complex transformations occurring in the atmosphere. Over 100 chemical reactions may potentially play a role in transforming SO<sub>2</sub> to sulfate. Unfortunately, the chemical transformation process cannot be completely evaluated at present, since important elements of many of the equations are not known. The importance of understanding the full ensemble of chemical reactions cannot currently be evaluated. A more pertinent question is: how accurately can a linear relationship approximate these reactions over the large time and space scales involved in *regionwide* emissions and deposition?

Clearly the total amount of sulfur emitted to the atmosphere is eventually deposited; thus, ultimately, reductions in deposition will approximate emissions reductions. (Deposition from natural sulfur sources, though small, would remain.) The crucial question is *where* sulfur deposition would be reduced—i.e., in about the same regions as predicted by the linear models, closer to emissions sources, or further away?

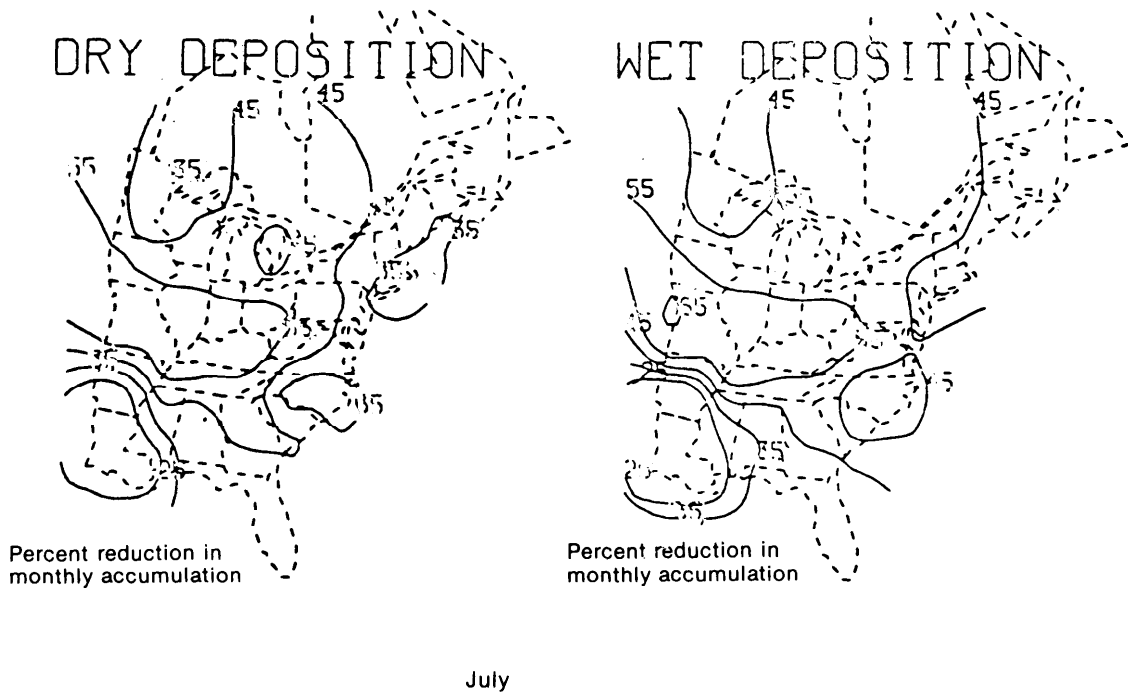
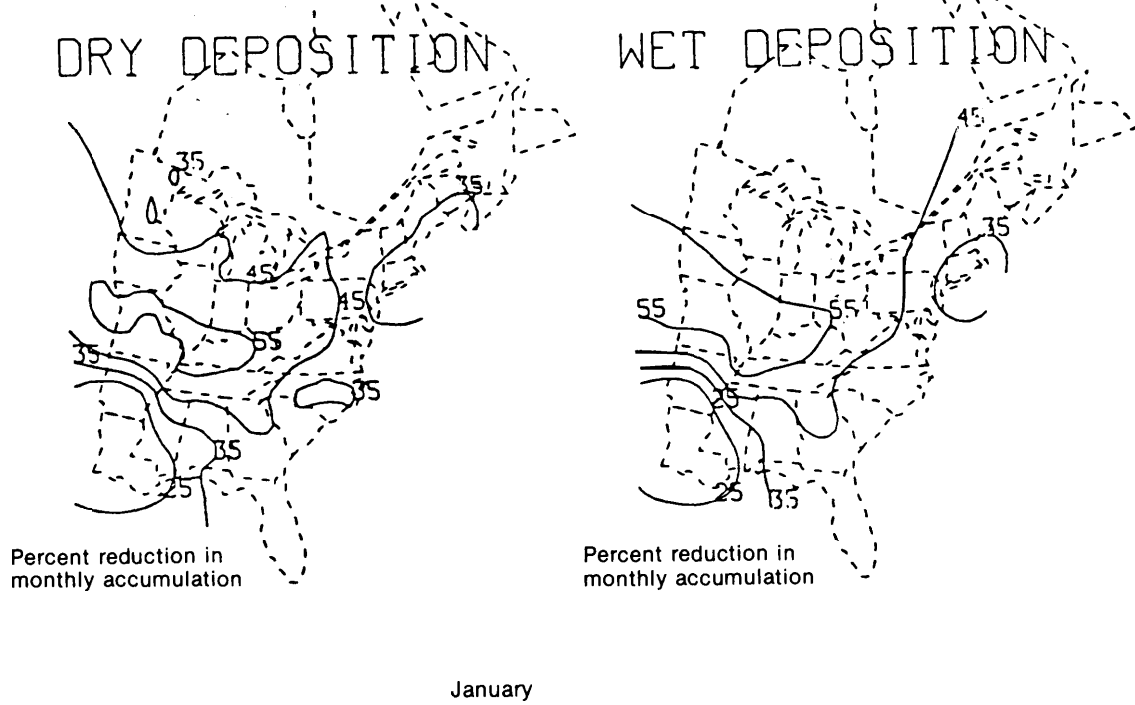
To examine the linearity of the chemical transformation system, a second model, capable of simulating the interactions of several pollutants that may affect sulfur deposition, was run for OTA.<sup>8</sup>

This model, developed by Rodhe, et al. (1981), allows a reasonable qualitative evaluation of pollutant interactions while remaining computationally tractable. The model's limited description of atmospheric mechanisms (e. g., mixing of pollutants with surrounding air) could affect its quantitative results. In addition, it does not incorporate the meteorology necessary to describe complex patterns of deposition. Nonetheless, it represents a useful step forward from linear modeling, by simulating chemical transformations that occur in the atmosphere, through a series of 19 chemical equations. The Rodhe model can provide a qualitative picture of how changes in other primary pollutant emissions—principally reactive hydrocarbons (RHC) and nitrogen oxides (NO<sub>x</sub>)—might affect downwind sulfur deposition.

Dry sulfur deposition depends highly on the concentration of SO<sub>2</sub> in the atmosphere. Changing the concentrations of RHC and NO<sub>x</sub> might alter the rate at which SO<sub>2</sub> is converted to sulfate, and change the reser-

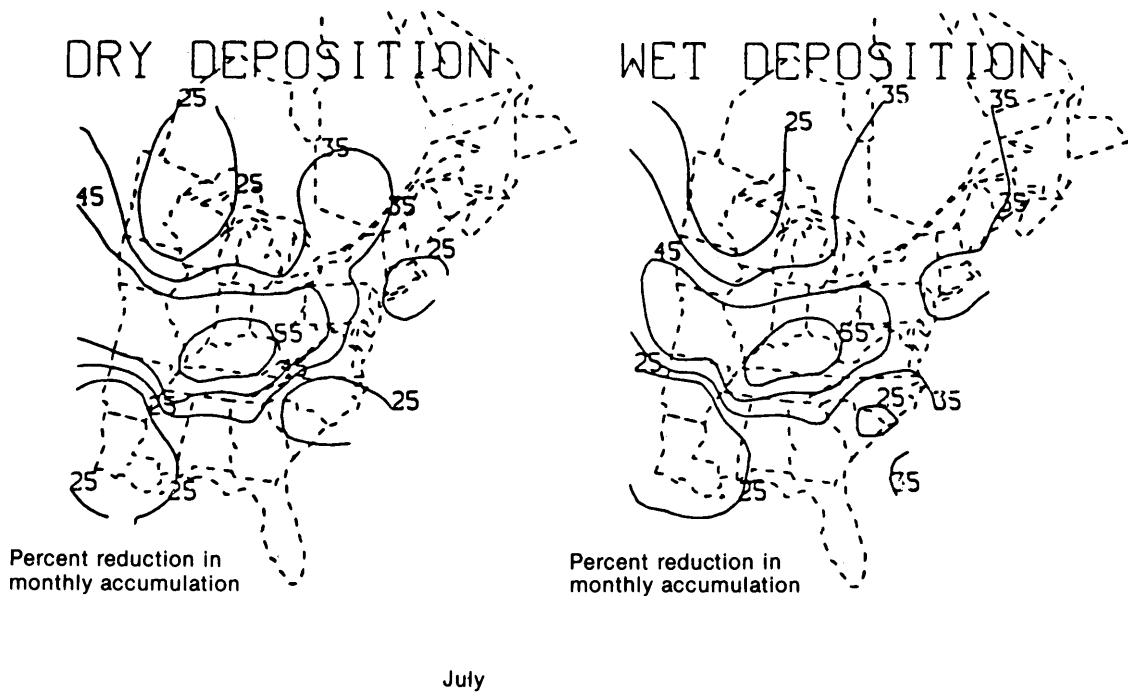
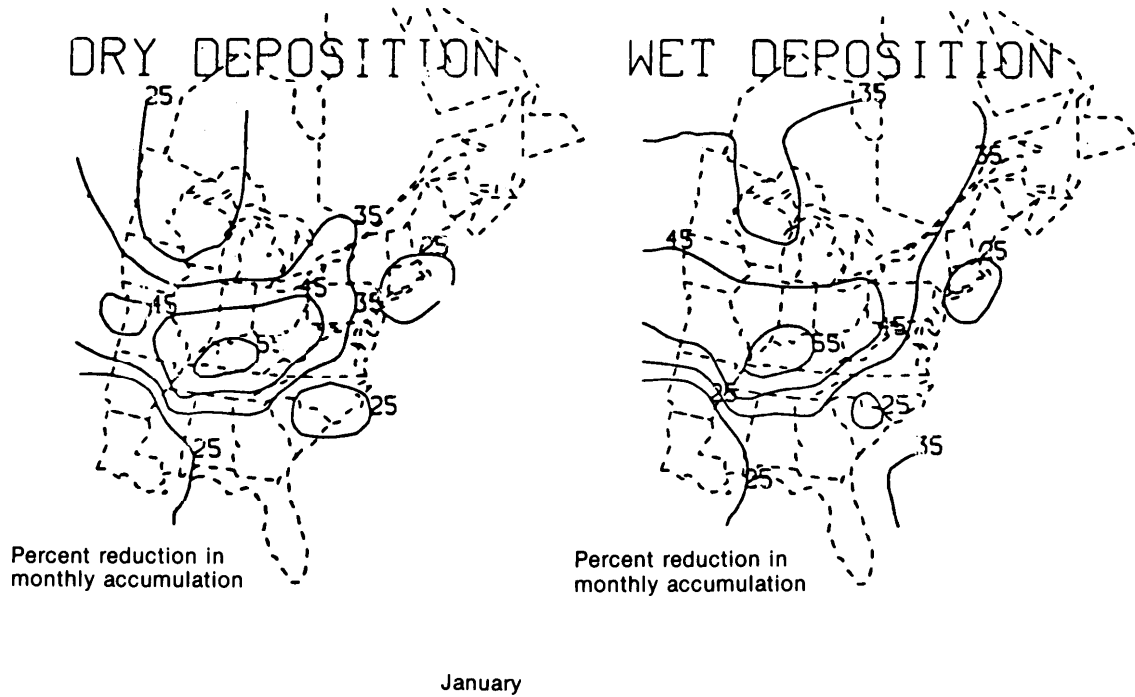
<sup>8</sup>Perry J Samson, "On the Linearity of Sulfur Dioxide to Sulfate Conversion in Regional-Scale Models, OTA contractor report, June 1982.

Figure C-13.—Estimated Deposition Following a 10-Million-Ton-per-Year Reduction in Sulfur Dioxide Emissions



NOTE: Emissions reductions allocated by S. 1706, 97th Congress. Estimates of deposition based on model results using OTA emissions analysis  
SOURCE: Jack Shannon, Argonne National Laboratory (ASTRAP model).

Figure C-14.- Estimated Deposition Following an 8-Million-Ton-per-Year Reduction in Sulfur Dioxide Emissions



NOTE: Emissions reductions allocated by S. 1706, 97th Congress. Estimates of deposition based on model results using OTA emissions analysis.  
SOURCE: Jack Shannon, Argonne National Laboratory (ASTRAP model).



voir of  $\text{SO}_2$  available for dry deposition. However, the model simulations indicate that dry sulfur deposition is fairly insensitive to the  $\text{NO}_x$  and RHC mixture in the atmosphere. Figure C-15 illustrates this by showing how much dry sulfur deposition changes per unit change of primary pollutant concentrations as a function of time downwind of a source region, as predicted by the Rodhe model. It shows that, of the three pollutants considered,  $\text{SO}_2$  has by far the greatest effect on dry sulfur deposition. In addition, the figure shows that, for pollutant travel times of less than 2 days, a given percentage reduction in emissions appears to reduce dry sulfur deposition comparably. Thus, since most dry deposition occurs within this time period, omitting co-pollutants and assuming a linear relationship in regional-scale models such as A STRAP may be a reasonable assumption for predicting changes in the dry sulfur component of deposition.

Wet sulfur deposition may respond to changes in pollutant emissions differently than dry deposition. As shown in figure C-16, changing the initial amounts of  $\text{SO}_2$  by 50 percent might result in roughly a 30-percent change in wet sulfur deposition. •One must keep in mind that these results are highly dependent on the model's approximation of chemical transformations in clouds. Because such processes cannot yet be simulated in detail, the accuracy of this result cannot be evaluated at this time.

However, like dry deposition, wet sulfur deposition is affected more by changes in  $\text{SO}_2$  concentration than by changes in other pollutant concentrations. Nonetheless, it should be noted that wet sulfur deposition is relatively sensitive to changes in RHC. Reducing RHC emissions could decrease wet sulfur deposition, although the effect would not be as pronounced as equivalent reductions in  $\text{SO}_2$ . The sensitivity of wet sulfur deposition to changes in  $\text{NO}_x$  concentrations is less pronounced, but in the opposite direction—i.e., reducing  $\text{NO}_x$  emissions might lead to a small increase in wet sulfur deposition. However, this increase might be offset by decreased dry sulfur deposition and reductions in nitrate deposition.

The Rodhe model is not intended to estimate pollutant deposition quantitatively. It was employed to evaluate pollutant interactions qualitatively and assess how they might affect dry and wet sulfur deposition. The results suggest that *wet* sulfur deposition is relatively sensitive to the mix of RHC and  $\text{NO}_x$ , while *dry* sulfur deposition is not.

\*This estimate is for: 1) relative, high initial concentrations of pollutants, such as those found in the Ohio River basin during pollution episodes, and 2) an "average" rainfall event (1 hour at 1 mm/hr) 24 hours downwind of the source.

An analysis performed by a committee of the National Research Council/National Academy of Sciences<sup>9</sup> (NAS) came to a similar conclusion. This committee slightly modified the Rodhe model used by OTA to reflect some recent laboratory results and to change some assumptions about mixing of chemicals in the atmosphere. Their results suggest that the role of  $\text{NO}_x$  and RHC are somewhat less important than indicated by the "worst-case" assumptions used by OTA.

Both the OTA and NAS analyses suggest that if reducing total sulfur deposition is a desired goal, reducing  $\text{SO}_2$  emissions would likely be an effective strategy. The NAS report concludes,

If we assume that all other factors, including meteorology, remain unchanged, the annual average concentration of sulfate in precipitation at a given site should be reduced in proportion to a reduction in  $\text{SO}_2$  and sulfate transported to that site from a source or region of sources. If ambient concentrations of  $\text{NO}_x$ , nonmethane hydrocarbons, and basic substances (such as ammonia and calcium carbonate) remain unchanged, a reduction in sulfate deposition will result in at least as great a reduction in the deposition of hydrogen ion.

#### **"Target" Deposition Rates To Protect Sensitive Resources**

The previous discussion considered only the extent to which decreasing  $\text{SO}_2$  emissions by 8 million and 10 million tons per year in the Eastern 31-State region would reduce total sulfur deposition. No attempt was made to relate these estimates to preventing potential damage to sensitive resources. At least three separate groups have estimated deposition "targets" —i.e., levels of deposition below which sensitive lakes and streams are not expected to further acidify. \* \*

The Impact Assessment Working Group established under the U.S.-Canada Memorandum of Intent on Transboundary Air Pollution suggested the first of these targets.<sup>10</sup> Both the U.S. and Canadian members agreed that for North America:

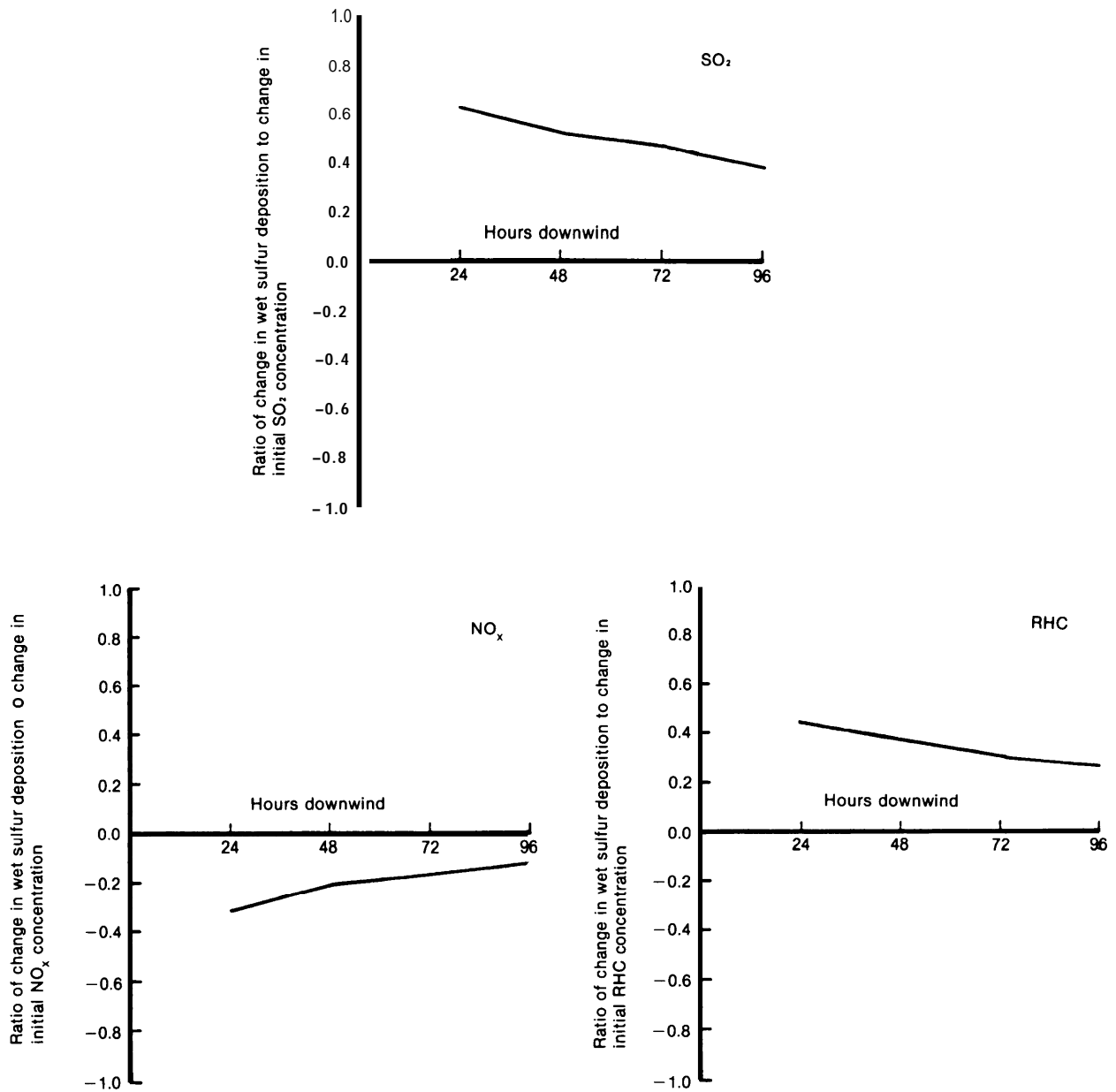
There have been no reported chemical or biological effects for regions currently receiving loadings of sulphate in precipitation at rates less than about 20 kg/ha-yr [kilograms per hectare per year]. Evidence of chemical change exists for some waters in regions currently, estimated or measured to be receiving about 20-30 kg/ha-yr sulphate in precipitation . . . Long-term chemical

<sup>9</sup>NRC /NAS, *Acid Deposition. Atmospheric Processes in Eastern North America*, National Academy Press, Washington, DC., 1983

• \*OTA estimates of the effects of changes in deposition on acid-altered aquatic resources are presented in appB. Rather than presenting specific deposition targets to prevent acidification of sensitive resources, the effects of three scenarios of changes in wet sulfur deposition are discussed.

<sup>10</sup>Impact Assessment, Work Group 1, U.S.-Canada Memorandum of Intent on Transboundary Air Pollution, Final Report, January 1983

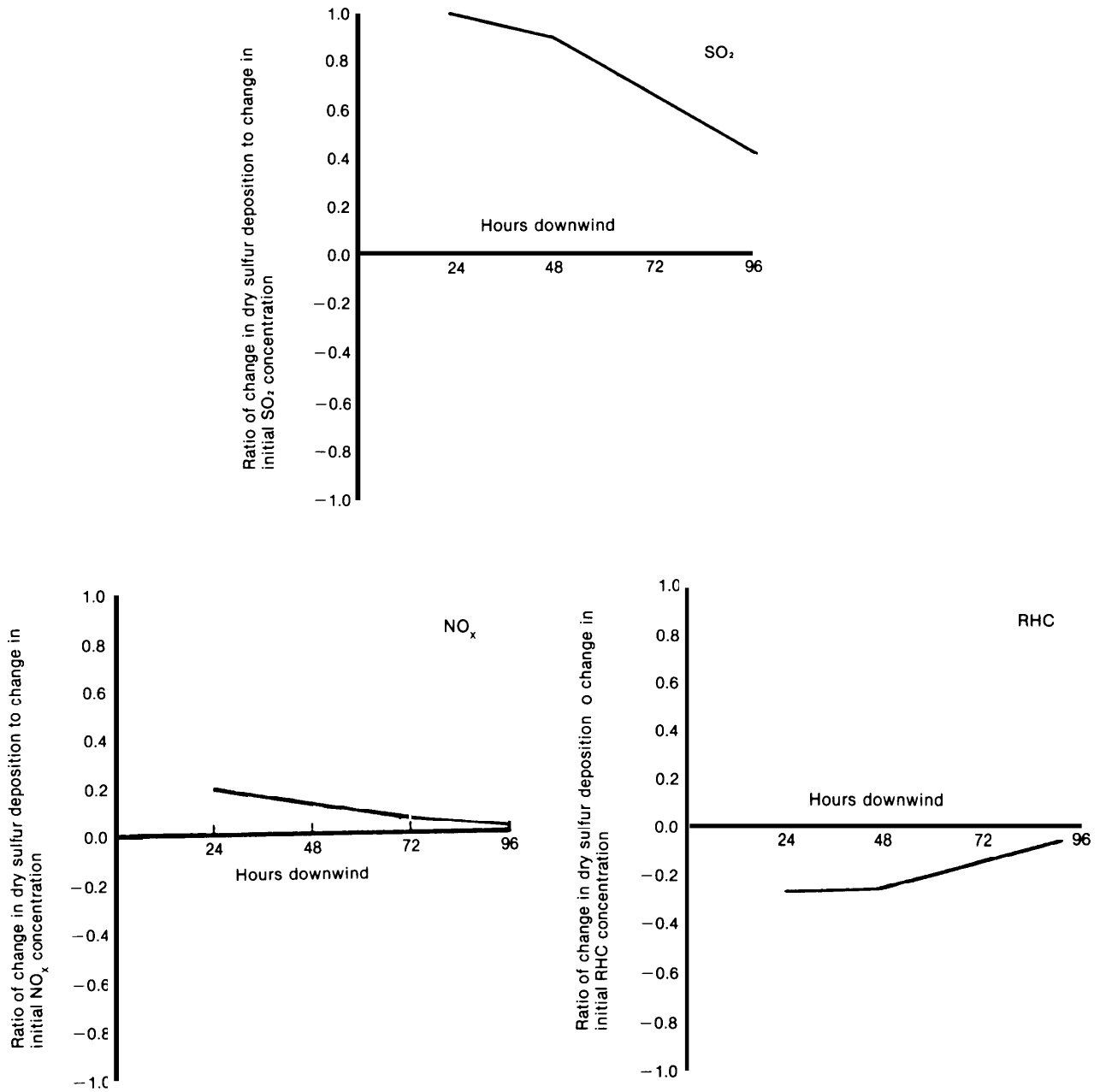
Figure C-15.—Changes in Wet Sulfur Deposition



The curves project the changes in potential *wet* sulfur deposition resulting from a unit change in the initial concentrations in the primary pollutants. Changes in deposition are plotted as a function of time downwind from the source region. These estimates assume a precipitation rate of 1 mm/hr at a given time.

SOURCE: Perry Samson, OTA contractor report, 1982.

Figure C-16.—Changes in Dry Sulfur Deposition



The curves project the changes in potential *dry* sulfur deposition resulting from a unit change in the initial concentrations in the primary pollutants. Changes in deposition are plotted as a function of time downwind from the source region.

SOURCE: Perry Samson, OTA contractor report, 1982.

and/or biological effects and short-term chemical effects have been observed in some low alkalinity surface waters experiencing loadings greater than about 30 kg/ha-yr. Based on these observations, the Canadian members of the group proposed that

. . . deposition of sulfate in precipitation be reduced to less than 20 kg/ha-yr [about 18 lbs/acre-yr] in order to protect all but the most sensitive aquatic ecosystems in Canada.

The U.S. members concluded that based on the current status of scientific understanding about the mechanisms that lead to surface water alteration, "it is not now possible to derive quantitative loading/effects relationships. \*

A recently published National Research Council/National Academy of Sciences (NAS) report uses the acidity of precipitation, rather than the concentration of sulfates, to specify a level that would protect sensitive freshwater ecosystems.<sup>11</sup> NAS states:

It is desirable to have precipitation with pH values no lower than 4.6 to 4.7 throughout such areas, the value at which rates of degradation are detectable by current survey methods. In the most seriously affected areas (average precipitation pH of 4.1 to 4.2), this would mean a reduction of 50 percent in deposited hydrogen ions,

Evans, Hendrey, Stensland, Johnson, and Francis presented a third estimate in a recent paper.<sup>12</sup> This group states:

For aquatic ecosystems, current research indicates that establishing a maximum permissible value for the volume weighted annual H<sup>+</sup> concentration of precipitation at 25 ueq/l may protect the most sensitive areas from permanent lake acidification. Such a standard would probably protect other systems as well.

This last estimate is quite similar to the National Academy estimate; a H<sup>+</sup> concentration of 25 ueq/l is equal to a pH of 4.6.

Maps of the decrease in *wet sulfur deposition* required to reach each of these targets can be derived from the measured deposition levels presented in figures C-4 to C-6. The three statements can be summarized into four plausible targets:

1. 20 kg/ha (42 meq/m<sup>2</sup>) wet sulfate,
2. average precipitation pH of 4.6 (25 ueq/l H<sup>+</sup>),
3. average precipitation pH of 4.7, and
4. 50-percent reduction in H<sup>+</sup> (hydrogen ion) deposited in rainfall.

It should be noted that these target values all use wet deposition as a surrogate for total (wet plus dry) deposition, because of the larger data base from which to draw comparisons to ecological effects.

Several assumptions are required for calculating the reductions in sulfate deposition necessary to reach the last three target values. First, if wet sulfate levels are reduced, a corresponding quantity of hydrogen ions (in absolute units, *not* percentage) must be eliminated to preserve the required charge balance. Second, one must assume that other ions, especially neutralizing ions such as calcium and ammonium, will remain at constant levels. Both assumptions are reasonable simplifications given the current chemical constituents in rainfall.

Figures C-17 to C-20 present maps of the percentage decrease in wet sulfur deposition required to reach the four deposition targets. These figures are derived from 1980 data—a year in which the annual precipitation was about average in most regions. Figure C-17 estimates regional wet sulfur-deposition reductions needed to meet the first target value (20 kg/ha wet sulfate). Figure C-17 shows that to reach this target, wet sulfur deposition would have to be reduced by about 50 percent in the areas of heaviest deposition, and by over 30 percent across broad regions of the Eastern United States.

Figures C-18 and C-19 show the wet sulfate-deposition reductions needed to meet target values of average rainfall pH of 4.6 and 4.7. The more stringent target of pH 4.7 requires greater than 70-percent decreases in areas of highest deposition. The less stringent target of pH 4.6 would require reductions about 10 percent smaller in the peak deposition areas; the required reduction drops by a larger percentage in areas of lower deposition.

Figure C-20 displays wet sulfate deposition reductions needed to meet target 4—a 50-percent reduction in hydrogen ion concentration in precipitation. The peak reductions required are on the order of 50 percent, with broad regions of the Eastern United States requiring wet sulfate reductions of 40 percent to reach this target.

Errors in sampling and chemical analysis lead to uncertainty about the position of the mapped lines showing required decreases of wet sulfate. These errors, which might be on the order of 10 percent, would translate into uncertainty about the position of the lines by about 50 to several hundred km,

Year-to-year weather variations could also shift the position of the lines on the maps. For comparison, projections of decreases in wet sulfur deposition necessary to meet target values 3 and 4 are shown in figures C-21 and C-22. These projections are based on 1979 data, rather than on 1980 data. For both target calculations, the 1979 data show *lower* required reductions, with the peak reductions shifted to the northeast. It is not possible to determine how much of the difference is attributable to sampling error, the smaller number of sampling stations operating in 1979, or differences in weather patterns.

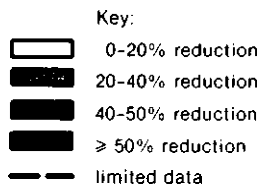
<sup>11</sup>NRC/NAS *Atmosphere-Biosphere Interactions: Toward a Better Understanding of the Ecological Consequences of Fossil Fuel Combustion*, National Academy Press, Washington, D. C., 1981.

<sup>12</sup>L. S. Evans, et al., "Acidic Deposition: Considerations for an Air Quality Standard," *Water, Air and Soil Pollution* 16, 1981, pp. 469-509.

Figure C-17.—Target Value: Wet Sulfate Loadings of 20 kg/ha-yr

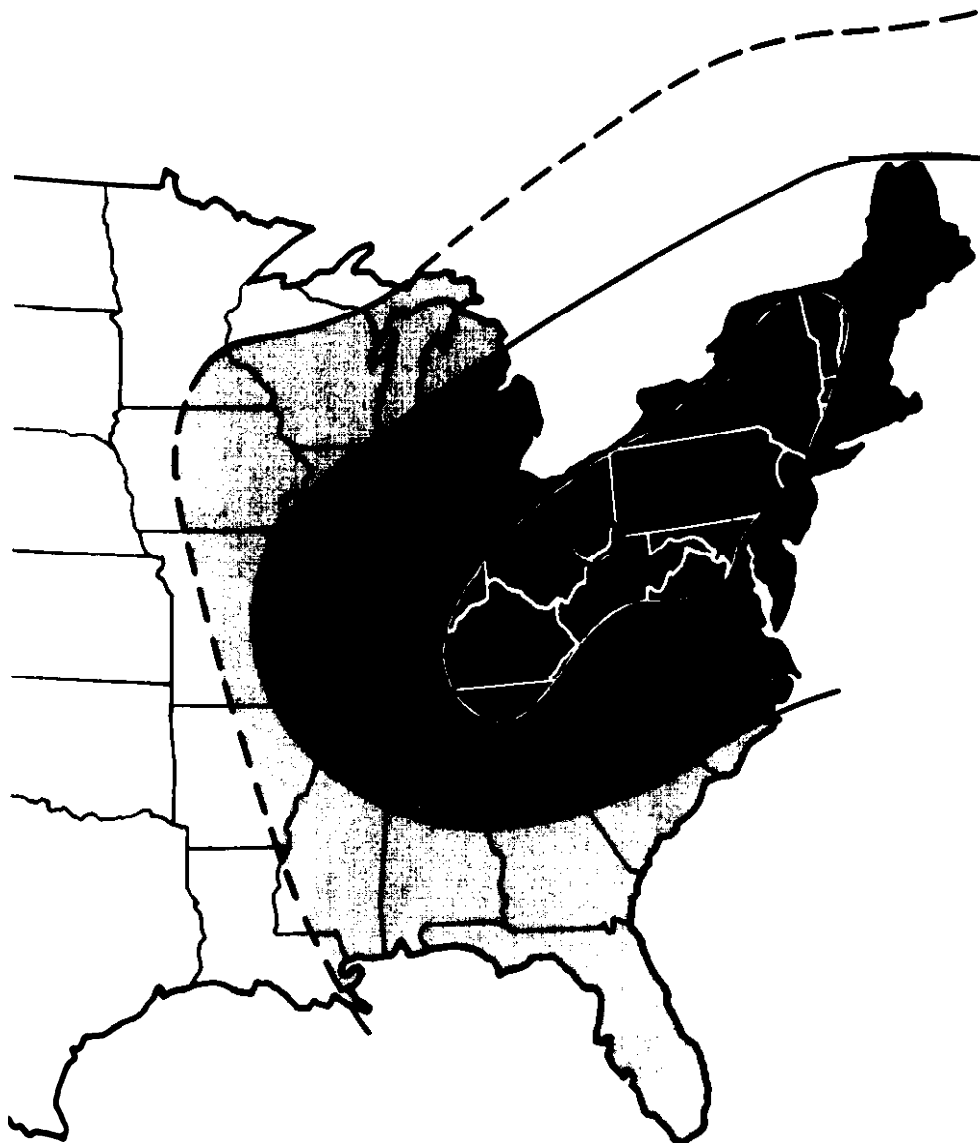


Estimated percent reduction in wet sulfate deposition necessary to reduce wet sulfate loadings to less than or equal to 20 kg/ha-yr (1980 data)



SOURCE: Office of Technology Assessment

Figure C-18.—Target Value: Average Precipitation pH of 4.6

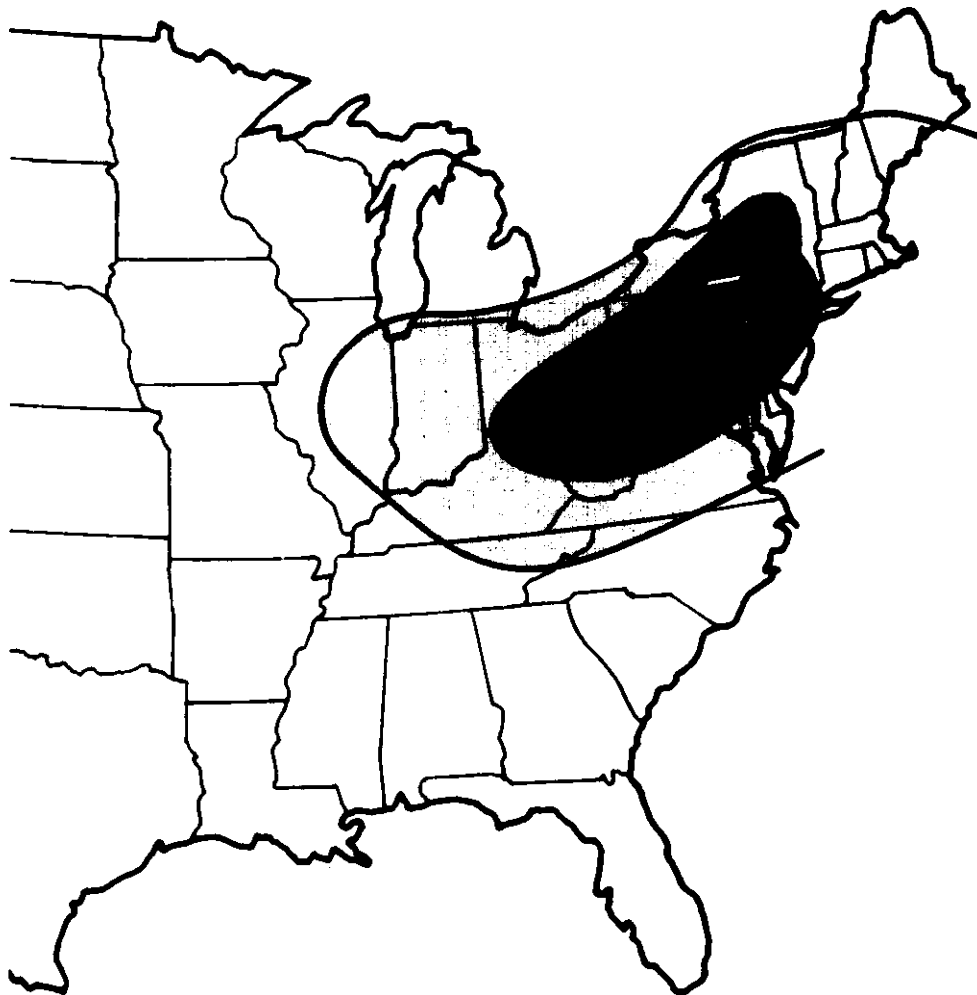


Estimated percent reduction in wet sulfate deposition necessary to increase pH to 4.6 or greater (1980 data).

- Key:
- 0-30% reduction
  - 30-50% reduction
  - 50-70% reduction
  - ≥ 70% reduction
  - - - limited data

SOURCE: Office of Technology Assessment.

Figure C-19.—Target Value: Average Precipitation pH of 4.7



Estimated percent reduction in wet sulfate deposition necessary to increase pH to 4.7 or greater (1980).

Key:

□ 50-70% reduction

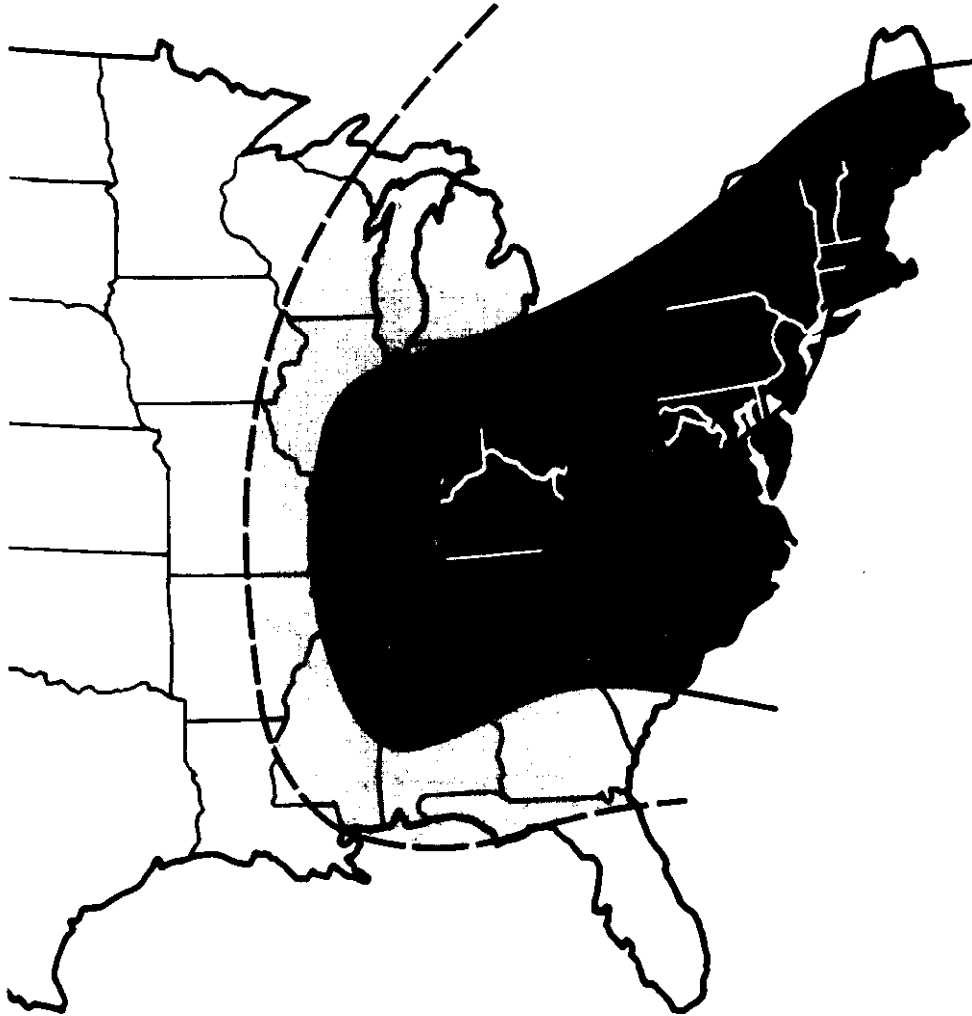
▨ 70-80% reduction

■ ≥ 80% reduction





- - - limited data

SOURCE: Office of Technology Assessment.

Figure C-20.—Target Value: 50% Reduction in Hydrogen Ion Deposition



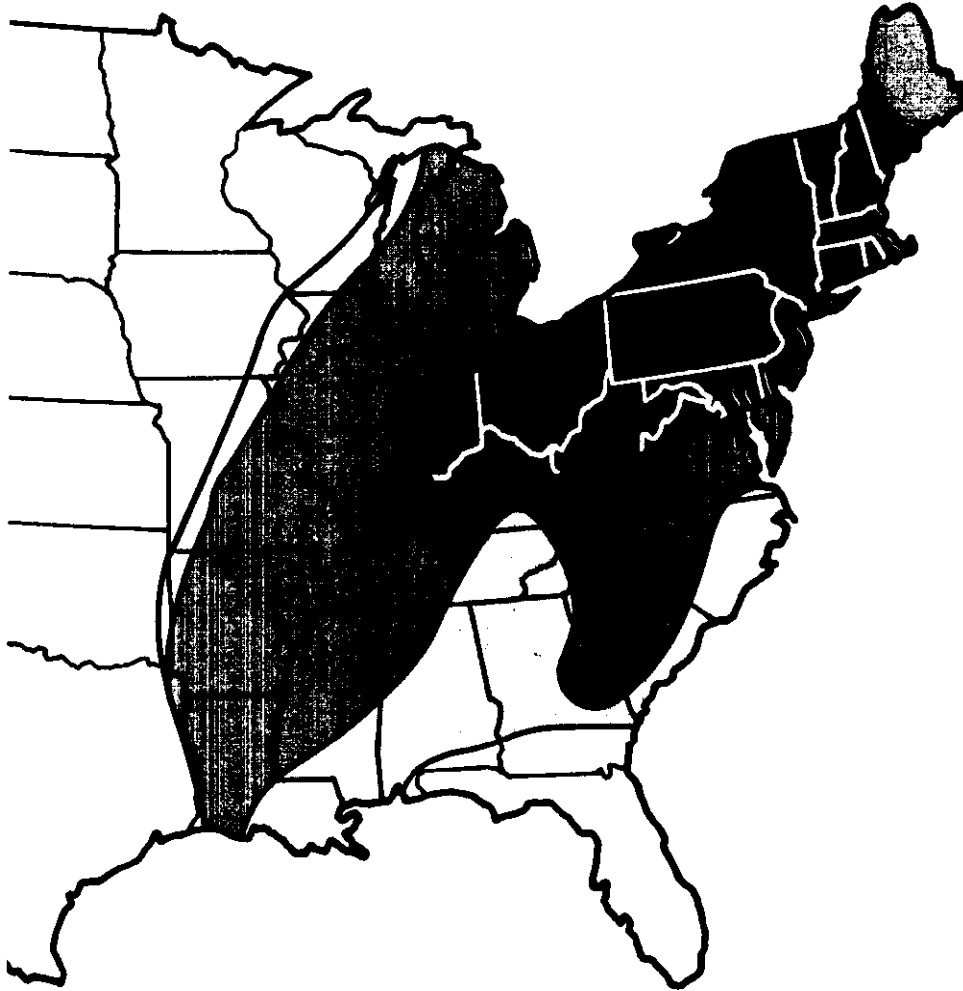
Estimated percent reduction in wet sulfate deposition so that hydrogen ion deposition is reduced by 50 percent (1980 data).

- Key:
-  30-40% reduction
  -  40-50% reduction
  -  ≥ 50% reduction
  -  limited data

SOURCE: Office of Technology Assessment.



Figure C-21.—Target Value: Average Precipitation pH of 4.7



This figure illustrates the estimated percent reduction in wet sulfate deposition necessary to reach the same target value as in fig. C-19—average precipitation pH of 4.7 or greater—however, the estimates are based on 1979 data.

Key:

1-20% reduction

20-40% reduction

> 40% reduction




SOURCE: Office of Technology Assessment.

Figure C-22.—Target Value: 50% Reduction in Hydrogen Ion Deposition



This figure illustrates the estimated percent reduction in wet sulfate deposition necessary to reach the same target value as in fig. C-20—i.e., a 50-percent reduction in hydrogen ion deposition—however, the estimates are based on 1979 data.

key:

-  1-20% reduction
-  20-40% reduction
-  > 40% reduction

SOURCE: Office of Technology Assessment.

As figures C- 17 through C-22 illustrate, the range of reductions required to reach each of the four targets—and the same target in two successive years—varies considerably. However, these figures permit some general *qualitative* statements to be made about the wet sulfate reductions needed to reach the suggested targets.

In regions of highest deposition, target deposition reductions range from 50 to 80 percent. For New England, deposition reductions required to reach the targets vary from about 30 to 70 percent. For sensitive areas south of the peak deposition region (e. g., the southern Appalachians in Tennessee), wet sulfur-deposition reductions needed to reach the targets are on the order of 20 to 50 percent. In the upper Midwest (e. g., the lake regions of Wisconsin and Minnesota), deposition would have to be reduced by 0 to 40 percent to reach the targets.

The previous section characterized the uncertainties inherent in using models to predict deposition reductions resulting from emissions reductions. Plausible model-based estimates of how much both wet and dry sulfur deposition might be reduced by an 8-million to 10-million-ton-per-year reduction in  $\text{SO}_2$  emissions were presented. Because the deposition targets are expressed

as decreases in wet sulfur deposition, and due to the large uncertainties in both sets of analyses, the deposition reductions that might result from reducing  $\text{SO}_2$  emissions can only be compared tentatively to the deposition reductions required to meet targets for protecting sensitive resources. In areas of highest deposition, for example, western Pennsylvania, the suggested target reductions to protect sensitive resources *might not* be achievable with an 8-million to 10-million-ton reduction in  $\text{SO}_2$  emissions; in areas of lower deposition, such as northern New England, the southern Appalachians and the upper Midwest, the target deposition reductions *might be* achievable with  $\text{SO}_2$  emissions reductions of this magnitude.

Thus, the uncertainties about reaching target deposition levels are greater in those areas that receive most of their sulfur deposition in wet form (e. g., the Adirondack Mountains). As discussed above, for a given level of  $\text{SO}_2$  emissions reductions, the reduction in wet deposition may be *less* than for dry deposition. Moreover, changes in wet sulfur deposition resulting from a given change in  $\text{SO}_2$  emissions may also depend on levels of such co-pollutants as  $\text{NO}_x$  and RHC.