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Project Summary

Historic Emissions of Sulfur and Nitrogen Oxides in the United States from 1900 to 1980

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Historic emissions of sulfur dioxide (SO_2) and nitrogen oxides (NO_x) were estimated for Task Group B, Manmade Sources, of the National Acid Precipitation Assessment Program for each state of the conterminous U.S. The emissions were estimated by individual source category on the state level from 1900 to 1980 for every fifth year and for 1978. The source categories included power plants, industrial boilers, industrial processes, commercial and residential heaters, natural gas pipelines, highway vehicles, off-highway diesel engines, and all other anthropogenic sources. These emissions were calculated from salient statistics indicative of fuel consumption or industrial output, estimations of average statewide fuel properties, and estimations of emission factors specific to each source category over time. The emission estimates were then aggregated to show the emission trends by state, region and all states combined. Total state emissions for each year were then estimated using an interpolation procedure based on national annual fuel consumption.

This Project Summary was developed by EPA's Air and Energy Engineering Research Laboratory, Research Triangle Park, NC, to announce key findings of the research project that is fully documented in a separate report of the same title (see Project Report ordering information at back).

Introduction

Sulfur oxides (SO_x) and nitrogen oxides (NO_x) are considered primary precursors of acidic precipitation. The anthropogenic emissions of these pollutants are suspected causes of many biological and chemical effects observed in recent years. Understanding the historic emission trends is important to understanding the development of acid-precipitation-related problems and causes of observed environmental effects.

Annual quantities of emissions of SO_x and NO_x are presented for each of the contiguous 48 states and the District of Columbia. Emissions of each pollutant were estimated for every fifth year from 1900 to 1980 and for 1978. The period from 1900 through 1980 was selected to allow study of early alkalinity measurements and also to allow comparison with the 1980 national emission inventories being developed under the National Acid Precipitation Assessment Program (NAPAP). Five-year intervals were selected to provide an indication of the emission trends sufficient for most effects studies and to develop a methodology that could be applied to all other years. The state level was selected because it provides the most complete and consistent body of information on a historic basis and collectively covers all geographic regions of the country.

For each state the estimates are based on the apparent annual consumption rate of fuels. The fuels include bituminous coal, anthracite, lignite, residual and distillate oils, natural gas, wood, gasoline, diesel fuel, and kerosene. The consumers of these fuels, which are also the emitters of SO_x and NO_x, are categorized as electric utilities, industrial boilers, commercial and residential furnaces, pipelines, highway vehicles, railroads, coke plants, smelters, vessels, or other major sources. Emissions were also estimated for industrial processes based on production rates, wildfires, and a miscellaneous source category. Collectively, these source categories account for all anthropogenic emissions in each state.

Method

Average emission rates for each study year were calculated for individual source categories for each state. The source categories are listed in Table 1 according to the type of fuel consumed. These categories represent all types of boilers, furnaces, engines, processes, and other anthropogenic sources. The basic steps involved in calculating state emissions are:

1. Obtain *state level* information on fuel use.
2. Allocate fuel quantity used by each source category.
3. Develop source category emission factors.
4. Determine fuel sulfur content by state for each category.
5. Calculate emissions, after emission controls.

The actual procedure varied somewhat depending on the usefulness and availability of information. It can generally be described in more detail for two time periods: (1) 1950 to 1980; and (2) 1900 to 1945.

Approach for 1950 to 1980

Fuel Consumption—For electric utilities, state consumption rates of fossil fuels were derived by individual power plant from the Bureau of Census, the American Petroleum Institute, the U.S. Department of Energy (DoE), and the National Coal Association. The consumption rates were determined according to boiler type. For all other categories, except smelters and miscellaneous sources, annual fuel consumption rates were obtained for the source category as a whole from various publications. When fuel consumption data were not available, other salient statistics (e.g., fuel sales, demand, distribution, shipments) were used. For the highway vehicles category, vehicle miles traveled were used for 1970 to 1980 because these provided a better estimate according to the mix of vehicle types. For earlier years, gasoline consumption by state was used. For the wildfire category, total forest area burned in each state was used. For smelters and miscellaneous industrial processes, estimates were based on either individual plant or state production rates. For 1950 to 1980, state-level fuel consumption data were available for most source categories.

Table 1. *Fuel Types and Emission Source Categories*

Bituminous Coal:	*Electric Utilities* *Industrial Boilers and Space Heaters* *Commercial and Residential Uses* *Steam Railroads* *Coke Plants*
Anthracite Coal:	*All Uses*
Residual Oil:	*Electric Utilities* *Industrial Boilers and Space Heaters* *Commercial and Residential Uses* *Vessels*
Distillate Oil:	*Electric Utilities* *Industrial Boilers and Space Heaters* *Commercial and Residential Heating* *Railroads* *Vessels*
Natural Gas:	*Electric Utilities* *Industrial Boilers and Space Heaters* *Pipeline Compression Stations* *Commercial and Residential Uses*
Wood:	*Electric Utilities* *Industrial Boilers and Space Heaters* *Commercial Heating* *Residential Wood Stoves and Fireplaces*
Gasoline and Diesel:	*Highway Vehicles* *Off-Highway Vehicles* *Vessels*
Other:	*Wildfires* *Cement Plants* *Copper, Lead, and Zinc Smelters* *Miscellaneous Industrial Processes* *Miscellaneous Other Sources*

Emission Factors—The state-level data were then multiplied by specially derived emission factors to yield estimates of uncontrolled emissions. First, the most recent emission factors were obtained for stationary and mobile sources reported by the EPA. These factors are based on actual emission tests of each type of combustion process or emission source represented in each source category. They are most appropriate when applied to a large number of sources (e.g., on the state level). Periodically, they are revised by the EPA to include new, additional, or improved test data.

The factors for each pollutant were then adjusted to represent each source category as a whole. This procedure involved mathematically weighting each factor according to the amount of fuel consumed by various types of boilers, furnaces, engines, processes, or other emission sources comprising the category. For highway vehicles, NO_x emission factors were state specific and were weighted according to the amount of urban and rural traffic, state elevation, vehicle mix, and pollution controls in use. These adjustments provided the most representative factors for 1970 to 1980 for which vehicle miles traveled were available. For earlier years, vehicle miles traveled are not available; the factors were based instead on gasoline consumption and on estimated average miles per gallon for both urban and rural traffic. These factors yield generally the same results as those in a DoE report on the trend in internal compression ratios of vehicle motors.

SO_2 emission factors were also weighted according to fuel consumption by individual emission sources within each category. However, these factors are more dependent on fuel properties than on combustion sources and include a fuel sulfur content variable. The emission factors account for the fraction of the fuel sulfur that would be emitted as uncontrolled emissions and the remaining fraction that would be captured in the solid residue. These fractions are determined on the basis of source emission tests and materials balance analysis involving various coal ranks that are commonly used by each source. The average statewide values of sulfur content of coal on the consumer level were obtained from the Bureau of Mines for 1965. For the earlier years, average statewide values were calculated from fuel distribution reports and information on fuel properties by originating district. The average values of sulfur content of fuel oils were obtained from information published by the Bureau of Mines and the U.S. DoE, Bartlesville Energy Technology Center, for domestically produced fuel oils.

Emission Controls—The amount of emissions controlled by certain control devices was then subtracted from each source category. This step pertained to SO_2 which is controlled by flue gas desulfurization systems at power plants and by-product sulfuric acid plants at smelters. Controls applied to sources of NO_x emissions have generally had little effect in reducing emissions through 1980.

Estimates were then compared with EPA national emission estimates, with the NAPAP emission inventory for 1980, and with the estimates of E. H. Pechan, *et al.* for electric utility emissions. This provided an indication of the precision of estimates for common years and a basis for establishing the precision for earlier years.

Approach for 1900 to 1945

For this period, state-level data on actual fuel consumption by source category were not always available, especially for the earlier years. Also, the method for collecting and reporting early data was not always consistent with the method for more recent years. Depending on the type of information found, one of three approaches was taken:

1. State-level data were used when available.
2. National data were apportioned to the states.
3. No estimates were made when state and national data were unavailable and when the emissions were so small as to be considered *negligible*.

These approaches help account for most of the early SO_2 emissions which were dominated by coal usage and for which consumption data are available either on the state or national level. Most NO_x emissions are also accounted for by this approach, but in terms of quantity are comparatively less than SO_2 because of the low consumption rates of fuel oils and natural gas in the early study years.

Assumptions

The same sulfur content values derived for 1955 were assumed to apply to the earlier years. This assumption was necessarily made because no evidence was found to suggest a general trend in sulfur content. Available information indicates that sulfur content of coal as mined did not change significantly, and most coal was consumed in or near the producing states. Analysis of coal distribution patterns also suggests little change compared to the changes in the middle or recent part of the century.

It was also assumed that the emission factors used for 1955 applied to earlier years. No evidence was found to suggest a change in either the emission characteristic of coal-fired sources or the population mix of types of boilers and furnaces. As research in historic emission patterns and trends continues, this general assumption may be replaced by specific state-level data if such data can be developed.

Aggregation of Emissions

Historic fuel consumption data were tabulated for each state according to source category and study year. Corresponding fuel sulfur content values were also tabulated for each source category according to state and year. A third tabulation contained the weighted emission factors for SO_2 and NO_x by source category and in some cases by state. These three matrices were multiplied to produce two new matrices, one for SO_2 emissions and one for NO_x emissions. The emissions of each state were then totaled by year to provide an estimate of overall national emission trends. They were also totaled by fuel type and by source category to show the effects of fuel switching and changes in consumer sectors. State emissions were also aggregated to show the trends in broad geographic regions of the country.

The national emissions were then analyzed by season and also by stack height ranges. For the seasonal analysis, the percentage distribution of the emissions by season was estimated for each major source category based on engineering judgment and known historic characteristics of each source category to give a general indication of the trend since 1900. The total national emissions of each source category were then multiplied by these percentages. As a result, the estimated seasonal emissions reflect both the trend in total emissions by source category and the general change in the seasonal distribution of emissions by source category.

For the release height analysis, the percentage distribution of national emissions was estimated for each source category according to four broad ranges of stack heights. In the case of electric utilities, individual power plant emissions and stack height data were used to determine the national distribution by height from 1950 to 1980. For earlier years and for other sources, the analysis was based on the general trend in the stack heights for the category as a whole. Both the seasonal and stack height analyses provide an approximate indication *only* of the trend on the national level.

Yearly Estimates

State total emissions for the intervening years were interpolated from the state emissions estimated for the study years and the annual national energy consumption reported by fuel type. The interpolation was performed individually for each major fuel category by state. For each intervening year, the emissions of each fuel category were then added.

Results for 1900 to 1980

National and regional trends of each pollutant are presented here by fuel type and source category.

Fuel Consumption, Overall and for Categories

Figure 1 shows the total mineral fuel consumption for the U.S. in terms of energy consumed by major source as obtained from the Bureau of Census. Total coal consumption has remained relatively constant over time since 1900 compared to the consumption of other fuels. Since 1960, coal consumption has steadily increased on the national level by 30 percent. Crude petroleum and natural gas consumption (the primary sources of NO_x emissions) has increased most rapidly since the 1930s. Wood and anthracite accounted for a large portion of the total energy consumption in the early part of the century but not in the latter part. Until 1930, per capita energy consumption remained nearly constant, declined during the Great Depression and has increased since, except during the oil shortage in the mid-1970s.

SO_2 Emissions, Overall and for Categories

Figure 2 is a plot of the total quantity of SO_2 emissions for the contiguous U.S. by fuel type and for each study year. (Note that the emission levels for consecutive study years are interconnected only to highlight the historic trend. They do not necessarily represent the trends between

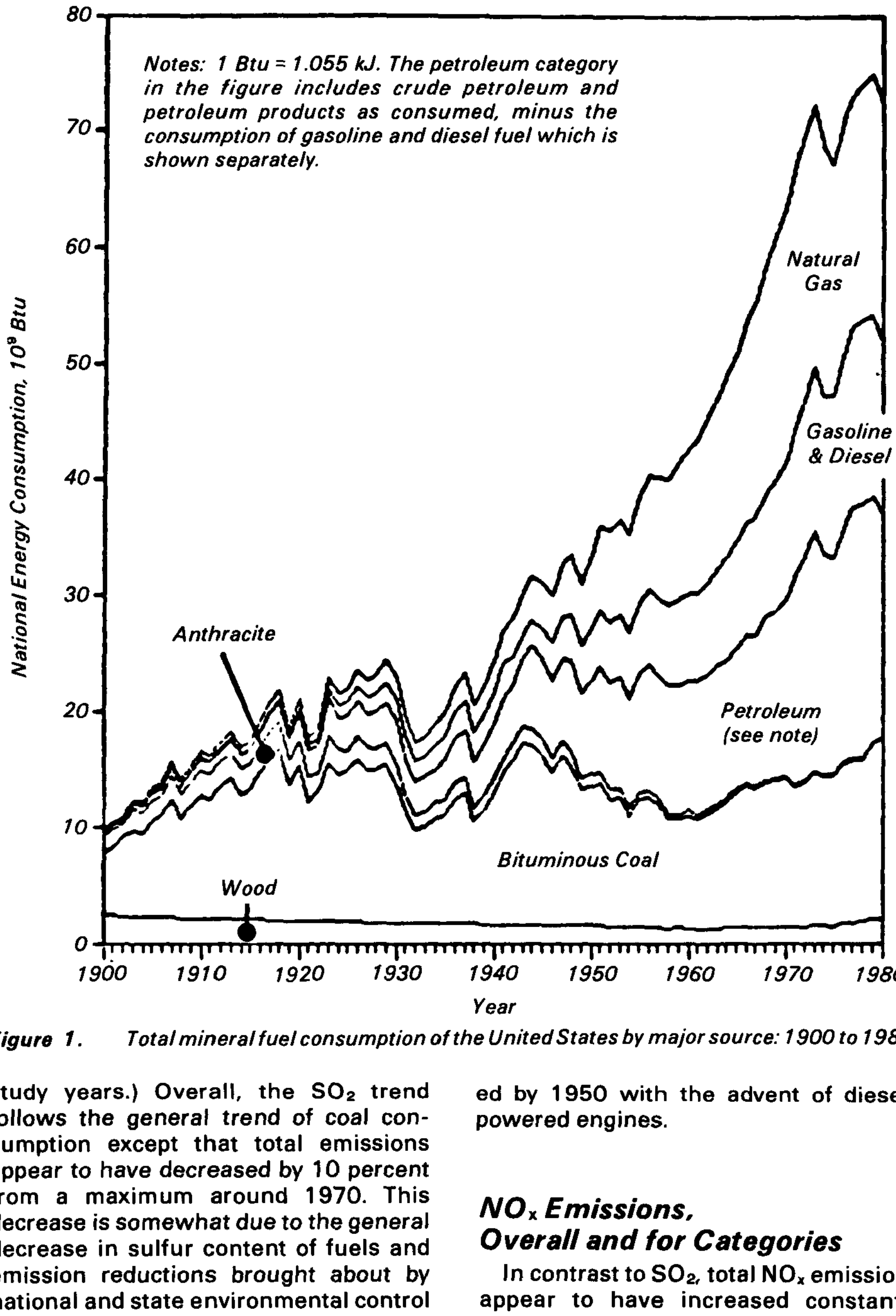

Figure 1. *Total mineral fuel consumption of the United States by major source: 1900 to 1980.*

study years.) Overall, the SO_2 trend follows the general trend of coal consumption except that total emissions appear to have decreased by 10 percent from a maximum around 1970. This decrease is somewhat due to the general decrease in sulfur content of fuels and emission reductions brought about by national and state environmental control regulations. Sulfur content has decreased to a large extent as a result of coal cleaning and mixing eastern coal with cleaner western coal, while in the early years coal was mostly burned as received from the nearest coal-producing district.

Figure 3 shows the overall trend by source category. This plot reflects the growth of major fuel-consuming sectors and changes in fuel demand. For example, electric utility emissions appear to have increased sharply by the 1950s and 60s. In contrast, SO_2 emissions from steam locomotives almost completely disappeared by 1950 with the advent of diesel-powered engines.

NO_x Emissions, Overall and for Categories

In contrast to SO_2, total NO_x emissions appear to have increased constantly throughout most of the study period as shown in Figure 4. The total quantity of emissions is plotted on the same scale as SO_2 to allow a direct comparison. This upward trend is primarily a result of greater use of natural gas and petroleum products and a conversion away from coal. Figure 5 shows that the increase is largely due to the growth in the number of highway vehicles, natural-gas-fired power plants, and many other sources related to a large extent to a growth in population and changes in technology and lifestyles.

Analysis by Region

The total state emissions were aggregated according to the Administrative Regions of the USEPA shown in Figure 6. These regions represent various broad geographic regions of the country. It should be noted that the regions vary in size and in the number of states and that these two factors will also affect the total regional emissions. By selecting a different combination of states other than these Federal regions, different emission trends may be shown. Recognizing this fact, the regions were selected to provide only a general indication of trends in various regions of the country.

The overall emission trend of each pollutant and the trend by category are plotted in Figure 7 for each region. These plots provide resolution of the national trend and allow the historic emission trends of each region to be compared. For example, Regions 3, 4, and 5 appear to have historically emitted more SO_2 than other regions in terms of total quantity. The total SO_2 emissions of Regions 1 and 2 combined have historically remained constant. In Region 6, NO_x emissions have increased more rapidly than in any other region due to the growth in the natural gas production industry and the number of pipeline compression stations. In all other regions, highway vehicles and electric utilities together have accounted for more than half the total NO_x in the past several decades.

In 1950, the regions east of the Mississippi River emitted 75 percent of the total national SO_2 emissions and 67 percent of the total NO_x emissions. In 1980, the eastern regions emitted 77 percent of the total SO_2 and 60 percent of the total NO_x. During this period, total national SO_2 emissions increased 140 percent while total NO_x emissions increased 280 percent, or twice as much. While most of the emissions have historically originated in the east, the western regions have begun to emit a greater share of the total national NO_x in recent years.

Analysis by Emission Release (Stack) Height

Analysis of emissions by release height (actual stack height) is important to studying the potential for long-range transport. Note that the *potential* for long-range transport increases with each higher range. This analysis does not include stack exit velocities or atmospheric mixing heights which are also important considerations. The analysis in this study suggests that more SO_2 emissions were released into the atmosphere from stacks above 240 ft* than from stacks below this height since about 1945. By 1980, approximately 30 percent of the SO_2 emissions were emitted above 480 ft, for example, compared to only 5 percent above this height in 1950. Not only have the percentages increased, but total national SO_2 emissions also increased and peaked around 1970. The percentage of the total SO_2 emissions released below 120 ft has generally decreased over the study period. The distribution of NO_x emissions has historically remained constant, although on the national level the total emissions have steadily increased. Approximately 60 percent of the total NO_x emissions in 1980 were released from ground level sources; predominantly from transportation sources.

Analysis of the electric utility category suggests that in the 1950s and 60s, most of the SO_2 and NO_x emissions from this category were released below 480 ft*—mostly between 240 and 480 ft. By 1980, about 50 percent of the total SO_2 emissions and 40 percent of the NO_x emissions from this source category were released above 480 ft as a result of the trend toward taller stacks. Since the emissions from electric utilities constitute a large portion of the total national emissions in recent years as shown in previous figures, they have a significant effect on the overall distribution of emissions by release height.

Conclusion

The current historic emissions data file presents the best estimates available on the state level. The emission trends of each state vary over time reflecting changes in a variety of economic and technological factors. While the national and regional scale emissions data provide general indications of trends, it is recommended that the reader refer to the state-specific estimates presented in the full report for studies of the historic relationship between emissions and environmental effects. These emission estimates can serve as the basis for future studies of the relationship between emissions and environmental effects associated with acid precipitation phenomena.

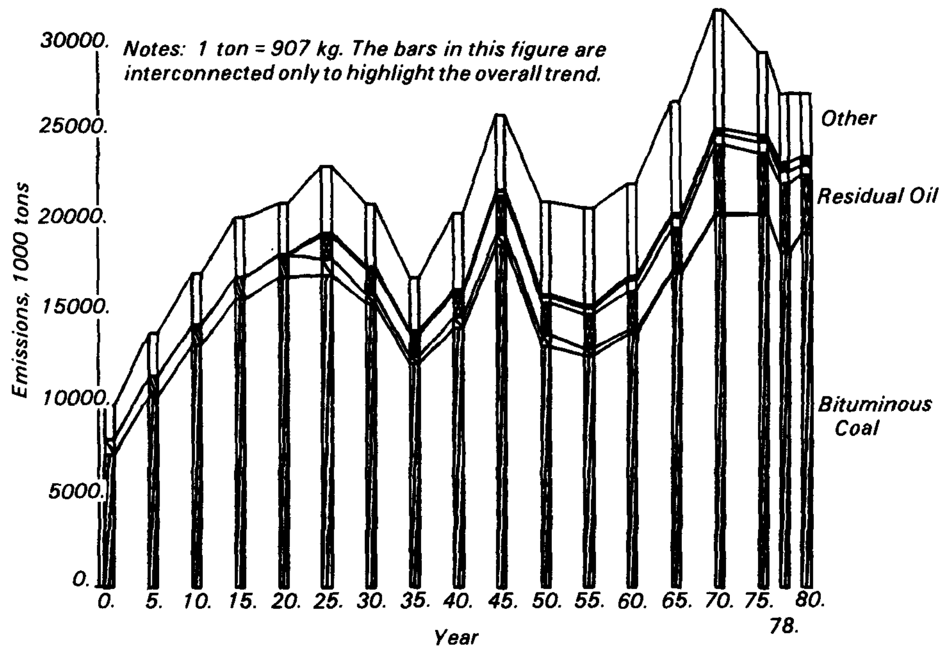

Figure 2. *Overall trend in SO_2 emissions from 1900 to 1980 for the U.S. and by fuel type for each study year.*

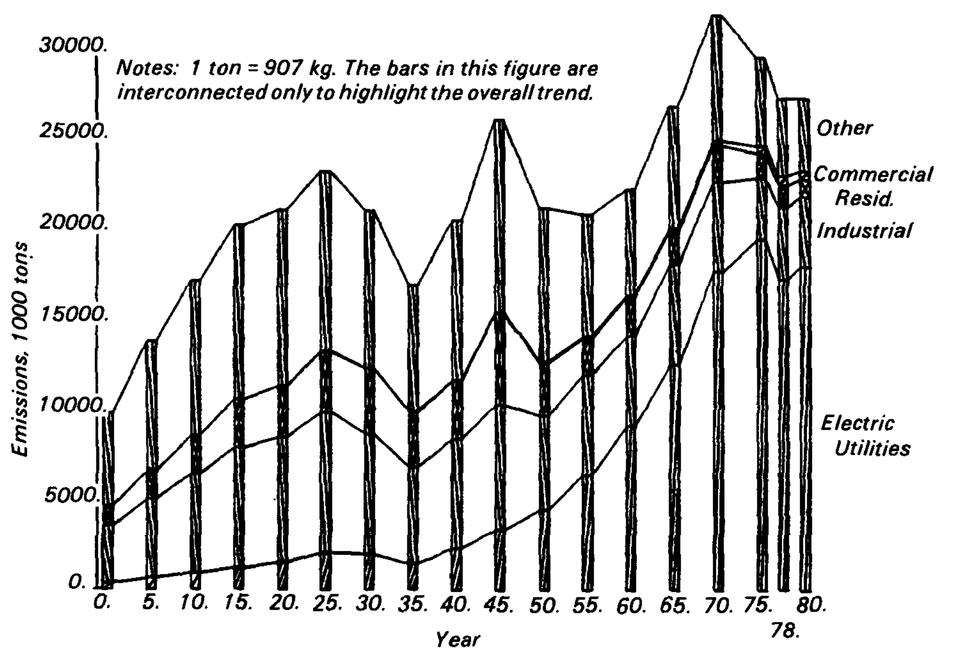

Figure 3. *Overall trend in SO_2 emissions from 1900 to 1980 for the U.S. and by source category for each study year.*

(*) 1 ft = 30.5 cm

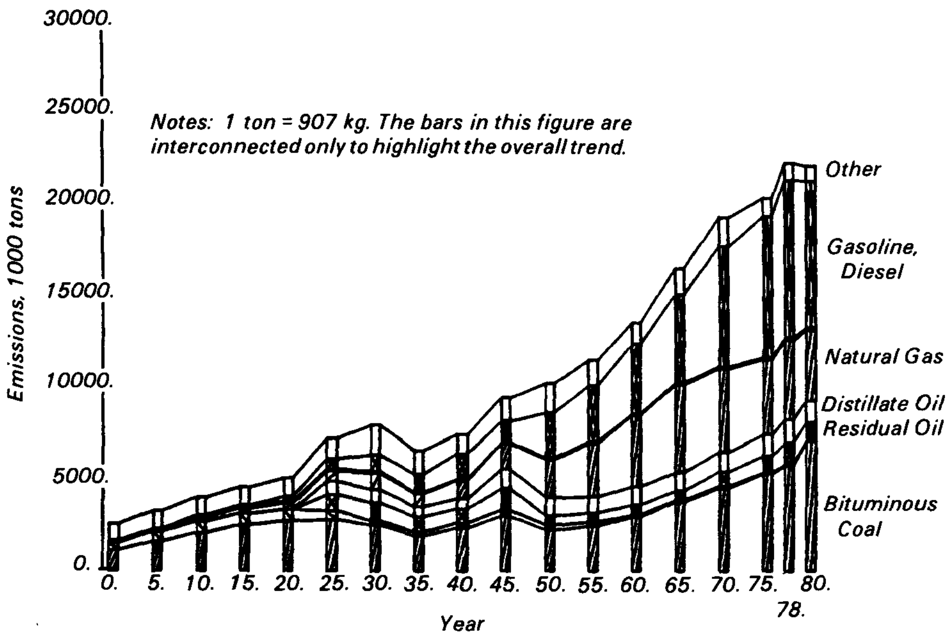

Figure 4. Overall trend in NO_x emissions from 1900 to 1980 for the U.S. and by fuel type for each study year.

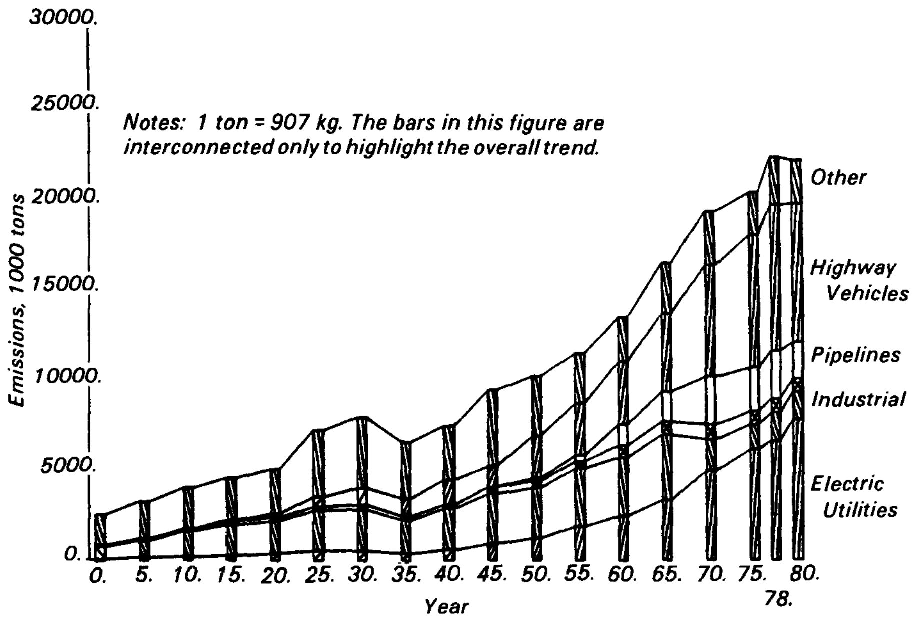

Figure 5. Overall trend in NO_x emissions from 1900 to 1980 for the U.S. and by source category for each study year.

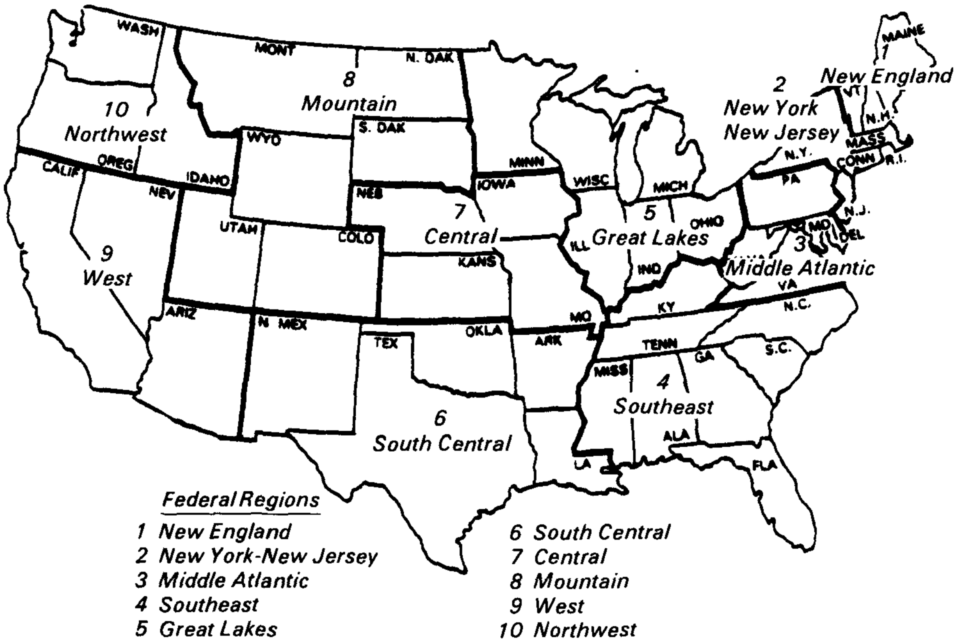

Figure 6. *Map of the EPA administrative regions (study did not include Alaska, Hawaii, and the Territories).*

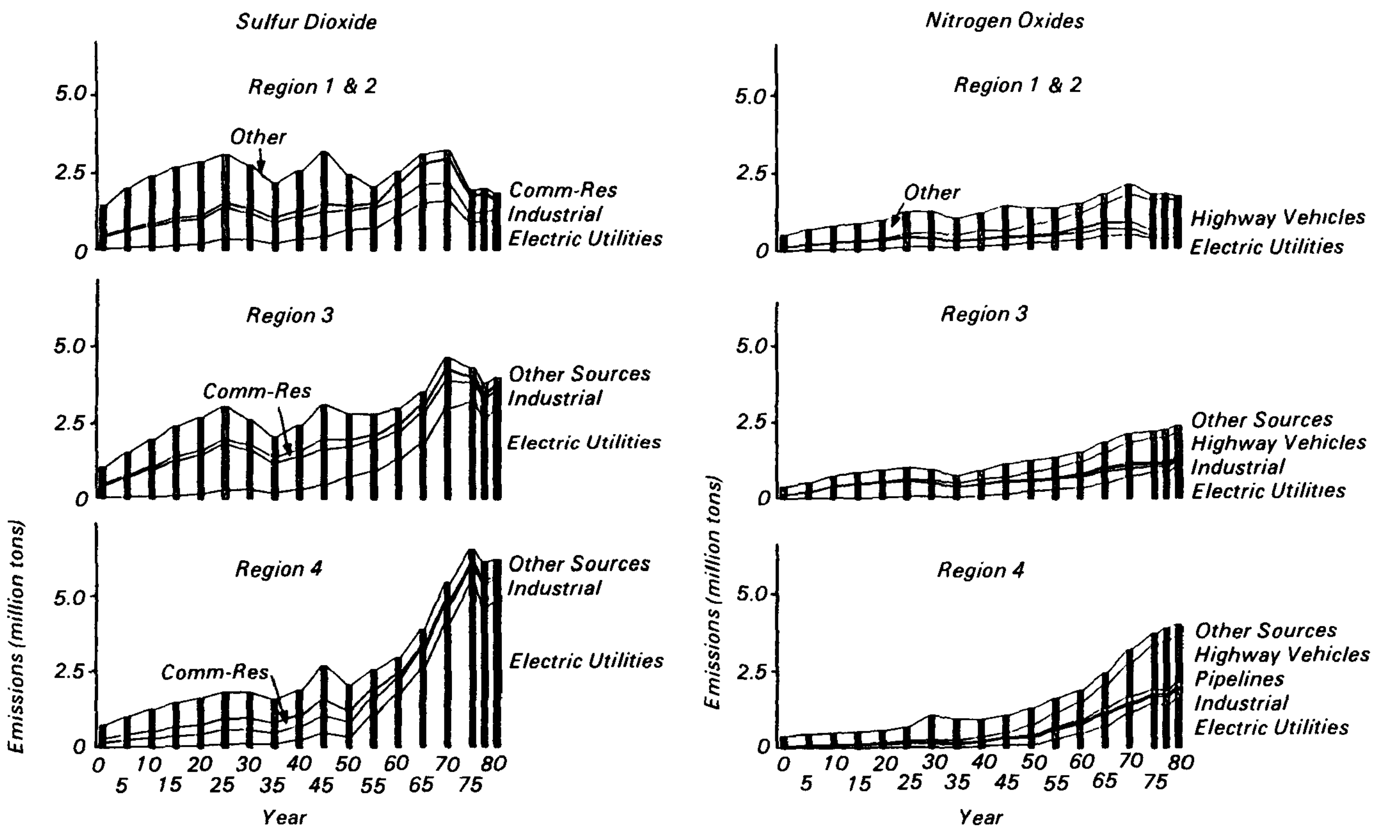

Figure 7. *Temporal changes in regional SO_2 and NO_x emissions by source category from 1900 to 1980. (Note: Emission estimates for years prior to 1950 may not account for all emissions due to incomplete data. Vol. II of full report has details on state level. The category "other" includes railroads, vessels, off-highway diesels, wildfires, smelters, and miscellaneous. 1 ton = 907 kg.)*

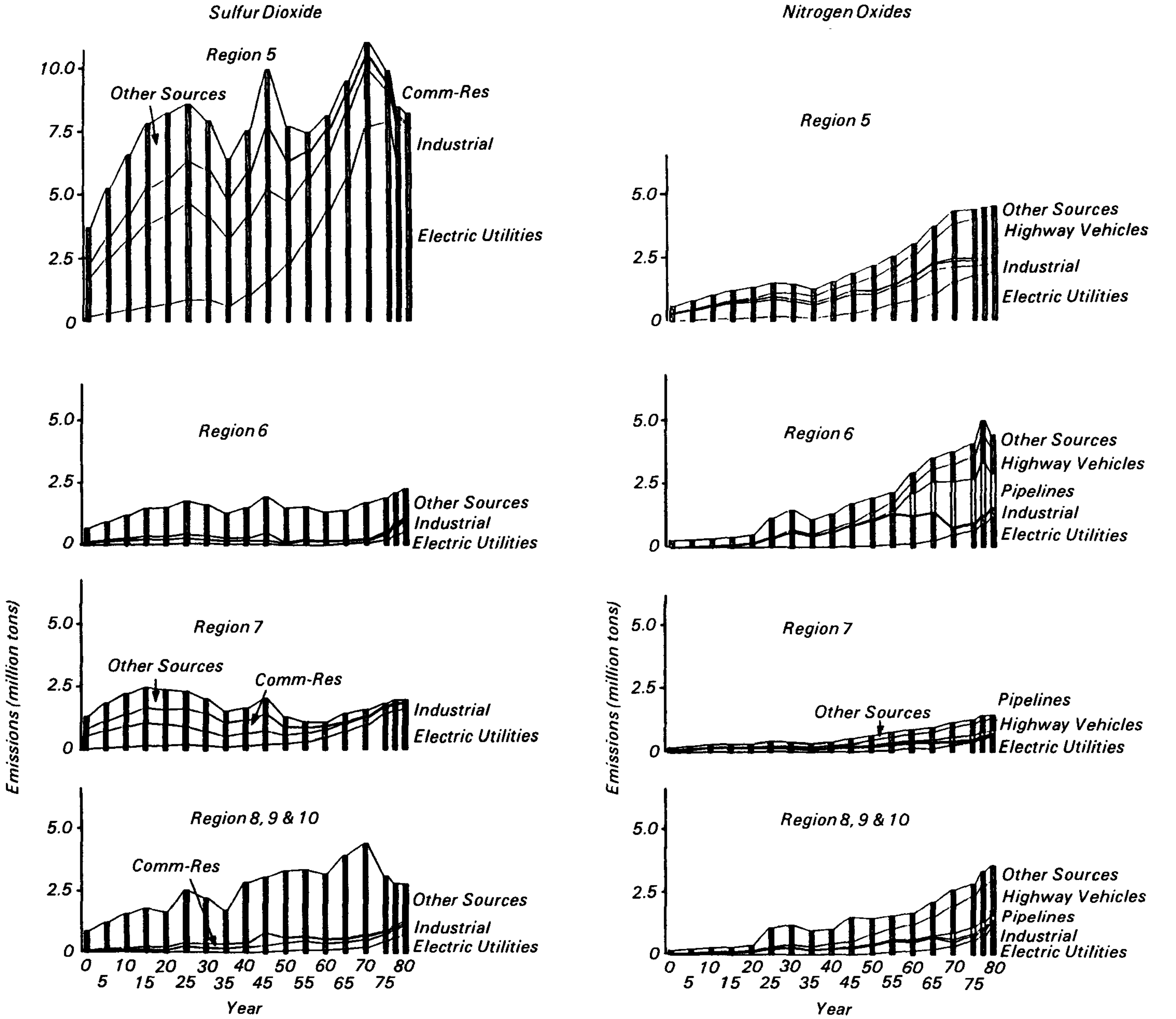

Figure 7. *(Continued) Temporal changes in regional SO_2 and NO_x emissions by source category from 1900 to 1980.*

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The complete report consists of two volumes, entitled "Historic Emissions of Sulfur and Nitrogen Oxides in the United States from 1900 to 1980:"
"Volume I. Results," (Order No. PB 85-191 195/AS; Cost: $13.00)
"Volume II. Date," (Order No. PB 85-191 203/AS; Cost: $46.00)
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