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EXECUTIVE SUMMARIES WORK GROUP REPORTS

UNITED STATES — CANADA

MEMORANDUM OF INTENT ON TRANSBOUNDARY AIR POLLUTION

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FEBRUARY 1983

EXECUTIVE SUMMARIES

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Submitted to the Coordinating Committee in fulfillment of the requirements of the Memorandum of Intent on Transboundary Air Pollution signed by Canada and the United States on August 5, 1980.

43-236-189

EXECUTIVE SUMMARY

WORK GROUP 1

SECTION 1

SUMMARY

1.1 INTRODUCTION

Wet and dry deposition of acidic substances and other pollutants are currently being observed over most of eastern North America. The Impact Assessment Work Group was charged with identifying and making an assessment of the key physical and biological consequences possibly related to these transboundary air pollutants.

During the Work Group's assessment of these effects it has been necessary to conduct the work along strictly disciplinary lines. Thus the presentation of our findings follows a sectoral approach (i.e., aquatic, terrestrial). While this approach has been useful for organizing and presenting our findings, it has also limited our consideration of the interactions which exist among these sectors. These effects do not occur in isolation.

The following sections summarize findings of the Work Group with respect to impacts on aquatic and terrestrial sectors of the biosphere, health and visibility, and man-made structures. There are also summary statements with regard to methodologies for estimates of economic benefits of controls, natural and material resource inventory, and liming.

1.2 AQUATIC ECOSYSTEM EFFECTS - CANADA

The potential effects from the deposition of acid and associated ions and compounds (sulphur dioxide, sulphate, nitrate, ammonia, and others) on water quality, and on the aquatic ecosystem, appear to be more fully quantified and understood than for terrestrial ecosystems. Data have been drawn from a number of study areas in eastern North America including Labrador, Newfoundland, Nova Scotia, New Brunswick, the southern part of the Canadian Shield in Quebec, and Ontario. Primary study areas in the U.S. are found in New Hampshire and southern Maine, Adirondack Park in New York, the Boundary Waters Canoe Area of Minnesota, and numerous lakes in north-central Wisconsin.

The findings and conclusions of the Work Group with respect to acidification effects are contained in the following statements:

Sulphuric acid has been identified as the dominant compound contributing to the long-term surface water acidification process. Nitric acid contributes to the acidity of precipitation, but is less important in eastern North America than sulphuric acid in long-term acidification of surface waters. Nitric acid contributes to pH depression of surface waters during periods of snowmelt and heavy rain runoff in some areas.

Studies of lakes in eastern North America have provided evidence that atmospheric deposition accounts for sulphate levels in excess of those expected from natural processes. In the absence of effects from mine drainage and industrial waste water, the symptoms of acidification (e.g., pH depressions of surface waters and loss of fish populations), have been observed only in lakes and rivers where the accompanying elevated concentrations of surface water sulphate (and nitrate in some cases) indicate atmospheric deposition of these ions. Land use changes, such as fires, logging, and housing developments have taken place in many areas with sensitive (low alkalinity) surface waters, but the symptoms of acidification have not been observed unless there is an accompanying increase in surface water sulphate concentrations. Nitrate concentrations also increase in some areas, especially during snowmelt.

In eastern Canada, the surface waters which have elevated excess sulphate occur in areas which have high atmospheric deposition of sulphate. All of the surface waters sampled in northeastern North America that have experienced loss of alkalinity also have elevated excess sulphate concentrations. In areas with less acidic deposition, loss of alkalinity in surface waters has not been observed. In Quebec, sulphate concentrations in surface waters decrease towards the east and north in parallel with deposition patterns. Sulphate concentrations are equal to or greater than the bicarbonate concentration in lakes in the southwest part of the Province. This indicates that the surface water chemistry has been altered by atmospheric sulphur deposition.

Observed Historical Changes

Sediments from lakes in Maine, Vermont, and New Hampshire indicate increased atmospheric acidic deposition has affected terrestrial and aquatic ecosystems as measured by changes in metal concentrations and diatom populations. It has been inferred from the sediment record that the rate of acidification of aquatic ecosystems has increased since the late 1800s as measured by declines in metals (zinc, copper, iron, calcium, magnesium and manganese) in the sediments. Conditions of low pH maintain metals in the water column, where they can be flushed out of the system before being deposited in the sediments. Diatom data are less complete, but they also indicate a statistically significant pH decline since the early 1900s.

In this report numerous historical chemistry records have been examined for waters not influenced by local urban or industrial discharges. Reviews have been conducted for 2 rivers in Newfoundland and 6 in Nova Scotia; 7 lakes in Nova Scotia and 3 in New Brunswick; 40 lakes in Adirondack Park, New York; 250 lakes in New England; 2 streams in New Jersey Pine Barrens; and 275 lakes in Wisconsin. Historical records which are available

from areas of soils and bedrock with a low potential to reduce acidity exposed to acidic deposition, show an increase in sulphate and corresponding decrease in alkalinity and pH. Areas of similar lithology and land use practices, but not receiving significant acidic deposition do not show similar losses of alkalinity.

Lakes in the Adirondack Mountain range have some of the lowest alkalinity values and are located in watersheds with a low potential to reduce acidity. They are located in the eastern U.S. in a zone receiving high acidic deposition (26-40 kg/ha.yr of sulphate in precipitation 1978-81). Historical data on fish and pH are available for 40 high elevation Adirondack lakes. In the 1930s, only 8% of these lakes had pH less than 5.0; 10% had no fish whereas in the 1970s, 48% had pH less than 5.0 and 52% had no fish. In some cases, entire fish communities consisting of brook trout, lake trout, white sucker, brown trout, and several cyprinid species apparently have been eliminated over the 40-year period. The New York Department of Environmental Conservation has concluded that at least 180 former brook trout ponds are acidic and no longer support brook trout. The relative contribution of natural and anthropogenic sources to acidification of these lakes is not known.

In New England, deposition of wet sulphate has been measured to be 17-40 kg/ha.yr. A study of 95 lakes for which there are historical pH data from the 1930s to the 1960s has indicated that 36% either had the same pH or higher while 64% now have lower pHs. For 56 lakes, a comparison of historical alkalinities to modern values indicated that 30% of the lakes had increased and 70% had decreased in alkalinity. Over the period of record, measured alkalinity values have decreased by an average of 100 μ eq/L. The lakes were small to medium size oligotrophic to mesotrophic with moderately to very transparent water, low to moderate concentrations of humic solute, low alkalinity and conductance and with moderately disturbed to pristine watersheds. For four rivers in Nova Scotia data from 1980-81 showed a decrease in bicarbonate, an increase in sulphate and hydrogen ion concentrations when compared to 1954-55 data.

Short-Term pH Depressions

While the rate of change of water quality of lakes (i.e., the time required for a lake to become acidified) is one of the least well-defined aspects of the acidification process, there is evidence that current acid loadings are damaging to fish populations and other biota due to short-term pH depressions following snowmelt and storm runoff. Both sulphate and nitrate are associated with short-term changes in water chemistry but in the majority of surveyed cases sulphate appears to be the larger contributor to the total acidity.

Short-term pH depressions, and elevated concentrations of metals, particularly aluminum, have been observed during periods of high infiltration or runoff. Metal accumulation in surface waters (Al, Mn, Fe, Zn, Cd, Cu, Pb, and Ni), first noted in streams and lakes of Scandinavia, also has been reported from such places as Hubbard Brook, the Adirondacks, and the Great Smoky Mountains of the U.S., and the southern Precambrian Shield area of Ontario, Canada. Artificial acidification of a lake in the Experimental Lakes Area of Ontario has also shown rapid mobilization of metals from lake sediments to the water column.

Data for 57 headwater streams in Muskoka-Haliburton show that 65% experience minimum pH values less than 5.5 and 26% have minimum pH values less than 4.5. Some inlet streams were observed to have pH values below 4.0 during spring snowmelt.

Data from intensive studies of 16 lakes in the Muskoka-Haliburton area of Ontario currently receiving about 23-29 kg/ha.yr sulphate in precipitation have shown that lakes which have summer alkalinity values up to about 40 $\mu\text{eq/L}$, experience pH depressions to values below about 5.5 during snowmelt. In Ontario and Quebec there are about 1.5 million lakes on the Precambrian Shield. In Ontario, of the 2,260 lakes sampled on the Precambrian Shield, 19% have alkalinities below 40 $\mu\text{eq/L}$. In the Shield area of Quebec, a 1981 survey of 162 lakes indicated 37% were extremely sensitive to acidification (CSI greater than 5.0), while 15% had summer pH values less than 5.0 (alkalinity less than 0).

A very large number of surface waters are being affected by acidic deposition, even though the total number of lakes and rivers in eastern North America which are known to have been acidified (alkalinity less than 0) by atmospheric acidic deposition is a relatively small percentage of the total aquatic resource.

Biological Effects

Detailed studies of watersheds have been carried out in sensitive regions of North America and Scandinavia under a range of sulphate deposition rates. The results of the studies conducted in North America are described below.

Observed changes in aquatic life have been both correlated with measured changes in the pH of water and compared for waters of different pH values. Differences have been documented in species composition and dominance and size of plankton communities in lakes of varying pH. Study results show that the number of species is lower in low pH lakes compared to lakes of higher pH. These alterations may have important implications for organisms higher in the food chain. Individual lakes often experience several symptoms of acidification at the same time. For example, in Ontario, Plastic Lake inlet streams have low pH

and high aluminum concentrations during spring runoff and extensive growth of filamentous green algae, and fish kills have been observed in Plastic Lake.

For those regions currently receiving loadings of sulphate in precipitation of less than 17 kg/ha.yr (Wisconsin, Minnesota and northwestern Ontario), there have been no observed detrimental chemical or biological effects.

For regions currently receiving between 20 and 30 kg/ha.yr sulphate in precipitation there is evidence of chemical alteration and acidification. In Nova Scotia rivers which currently have pH less than 5 there have been salmon population reductions as documented by 40 years of catch records. Fish stocks have remained viable in adjacent rivers with pH values presently greater than 5. Water chemistry records (1954-55 to 1980-81) have indicated a decline in pH to values presently less than 5 for other rivers in the same area. In Maine there is evidence of pH declines over time and loss of alkalinity from surface waters. In Muskoka-Haliburton there is historical evidence of loss of alkalinity for one study lake and there is documentation of pH depressions in all study lakes and streams with low alkalinity. Fish kills were observed in the shore zone of a study lake during spring melt. In the Algoma region there are elevated sulphate and aluminum levels in some headwater lakes.

For regions currently experiencing loading greater than 30 kg/ha.yr there are documented long-term chemical and/or biological effects and short-term chemical effects in sensitive (low alkalinity) surface waters.

In the Adirondack Mountains of New York, comparison of data from the 1930s with recent surveys has shown that some more lakes have been acidified. Fish populations have been lost from 180 lakes. Elevated aluminum concentrations in surface waters have been associated with low pH and survival of stocked trout is reduced by the aluminum.

In the Hubbard Brook study area in New Hampshire where the influx of chemicals is limited principally to precipitation and dry deposition there are pH depressions in streams during snowmelt of 1 to 2 units. Elevated levels of aluminum were observed in headwater streams.

Many species of frogs, toads and salamanders breed in temporary pools formed by the mixture of spring rains and snowmelt. Such pools are subjected to pH depression. Embryonic deformities and mortalities in the yellow spotted salamander which breeds in temporary meltwater pools have been observed in New York State where the acidity of the meltwater pools was 1.5 pH units lower than that of nearby permanent ponds. Population densities of

the bullfrog and woodfrog were reduced in acidic streams and ponds in Ontario.

A lake acidification experiment in northwestern Ontario clearly shows that alterations to aquatic food chains begin at pH values slightly below 6.0. The remarkable agreement between these whole-lake experiments and observational studies in Scandinavia and eastern North America provides strong evidence that the observed declines in fisheries are caused by acidification and not by other ecological stresses.

Extent of Effects

The terrestrial mapping analysis for eastern Canada supported by surface water chemistry has demonstrated that the watersheds of sensitive (low alkalinity) aquatic ecosystems where effects have been observed have a low potential to reduce acidity and are representative, in terms of soil and geological characteristics, of much larger areas of eastern Canada.

Similarly, using related but different criteria, maps have been developed which characterize considerable areas of the northeastern United States as having low potential to reduce acidity. Therefore, there is reason to expect that there are sensitive surface waters in these other areas which would experience similar effects if subjected to deposition rates comparable to those in the study areas. However, quantification of the number of lakes and rivers susceptible to acidification in both countries will require validation of the terrestrial mapping methodologies and increased information on the chemistry of lakes and streams.

The present empirical evidence covers a broad spectrum of physical and climatological conditions across northeastern North America and therefore provides a reasonable basis on which to make judgements on potential loading effect relationships. However the data do have some deficiencies. More data on historical trends of deposition and associated chemical and biological characteristics would improve our understanding of long-term rates and effects of acidification. In addition, a better understanding of all the mechanisms involved in the acidification process will enhance our ability to estimate loading/response relationships precisely. Therefore any estimates of loading/response relationships should be strengthened in the light of new scientific information as it becomes available.

Target Loadings

Sulphate in precipitation has been used as a surrogate for total acid loading. Sulphate in precipitation can be reliably

measured. It is recognized that dry deposition of sulphate and sulphur dioxide, and the wet and dry deposition of nitrogen oxides, nitric acid, particulate nitrate and ammonia, as well as other compounds also contribute to acidic deposition. Based on documented effects, wet and dry deposition of sulphur compounds dominate in long-term acidification.

Sulphur deposition also predominates in the majority of cases surveyed involving short-term pH depressions and associated effects. Insufficient data are available to relate nitrate deposition to short-term water quality effects. Therefore, we are unable to determine a nitrate dose-response relationship.

The models, which are based on theory, that have been considered, permit a quantification of the target loadings in terms of geochemical basin sensitivity. Although these models require further validation, the derived loading estimates are generally supportive of the empirical observations for the study areas discussed above.

Based on the results of the empirical studies, interpretation of long-term water quality data, studies of sediment cores and models that have been reviewed, we conclude that acidic deposition has caused long-term and short-term acidification of sensitive (low alkalinity) surface waters in Canada and the U.S. The Work Group concludes on the basis of our understanding of the acidification process that reductions from present levels of total sulphur deposition in some areas would reduce further damage to sensitive (low alkalinity) surface waters and would lead to eventual recovery of those waters that have already been altered chemically or biologically. Loss of genetic stock would not be reversible.

The Canadian members of the Work Group propose that present deposition of sulphate in precipitation be reduced to less than 20 kg/ha.yr in order to protect all but the most sensitive aquatic ecosystems in Canada. In those areas where there is a high potential to reduce acidity and surface alkalinity is generally greater than 200 $\mu\text{eq/L}$, the Canadian members recognize that a higher loading rate is acceptable.

As loading reductions take place and additional information is gathered on precipitation, surface water chemistry and watershed response, it may be possible to refine regional loading requirements.

1.2 AQUATIC ECOSYSTEM EFFECTS - UNITED STATES

Acidic deposition has been reported in the literature as a cause of both long-term and short-term episodic depressions in pH and loss in alkalinity in some lakes and streams in the U.S. and Canada.

Elevated concentrations of toxic elements, such as aluminum, and biological effects including losses in fish populations have been reported to accompany some of these pH depressions. In most of the reported cases, clear relationships were not established between acidic deposition and observed effects. Conclusions are based on an understanding of the acidification process although mechanisms which control this process are often not completely understood.

The following summary statements are observations reported to be occurring in areas receiving acidic deposition.

Both sulphuric and nitric acid contribute to the acidity of precipitation. It appears, however, that sulphuric acid contributes more to long-term acidification of surface waters than does nitric acid. Nitric acid can contribute to pH depression of surface waters during periods of snowmelt and heavy rain runoff in some areas. Studies of lakes in eastern North America indicate that atmospheric deposition accounts for sulphate levels in some waters in excess of those expected from natural processes. Lake study areas are located in Labrador, Newfoundland, Nova Scotia, New Brunswick, the southern part of the Canadian Shield in Quebec, and in eight regions of Ontario. Primary study areas in the U.S. are found in New Hampshire and southern Maine, Adirondack Park in New York, the Boundary Waters Canoe Area of Minnesota, and numerous lakes in north-central Wisconsin.

There is evidence of long-term reductions of pH and alkalinity and other water quality changes for some low alkalinity surface waters. The rate of change of pH and alkalinity in lakes is one of the least well defined aspects of the acidification process. However, there is evidence of short-term pH depressions in some waters following high runoff from snowmelt and storm activity. Both sulphate and nitrate are associated with short-term changes in water chemistry but, in the majority of surveyed cases, sulphate appears to be the larger contributor to total acidity.

Short-term pH depressions and elevated concentrations of metals, particularly aluminum, iron, zinc, and manganese have been observed during periods of high runoff. Metal mobilization from some watersheds, first noted in streams and lakes of Scandinavia, also has been reported from such places as Hubbard Brook, the Adirondacks, and the Great Smokey Mountains of the U.S., and Sudbury, Muskoka, and Plastic Lake in Ontario, Canada. Artificial acidification of a lake in the Experimental Lakes Area of Ontario has shown mobilization of metals from lake sediments to the water column.

Sediments from lakes in Maine, Vermont, and New Hampshire suggest increased acidity in aquatic ecosystems. It has been inferred from declines in metals (zinc, copper, iron, calcium, magnesium and manganese) in the sediments that the acidity of the water increased since the late 1800s. Low pH maintains

metals in the water column, where they can be flushed out of the system before being deposited in the sediments. Diatom data are less complete, but they also indicate a pH decline since the early 1900s.

There are few historical records of chemistry of low alkalinity waters not influenced by local urban or industrial discharges (i.e., 6 rivers in Nova Scotia; 7 lakes in Nova Scotia and 3 in New Brunswick; 40 lakes in Adirondack Park, New York; 250 lakes in New England; 2 streams in the New Jersey Pine Barrens; 270 lakes in Wisconsin). The above locations are exposed to various levels of acidic deposition. Some surface waters in these areas have shown a decrease in alkalinity and/or pH. In Wisconsin, however, most lakes surveyed had increased in alkalinity and pH.

The total number of lakes and rivers in eastern North America that are thought to have been acidified by acidic deposition is a very small percentage of the total aquatic resource. In the absence of effects from mine drainage and industrial waste water, the symptoms of acidification (e.g., long-term pH declines and/or short-term pH depressions of surface waters with loss of fish populations) have been observed only in clearwater lakes and streams with accompanying elevated concentrations of sulphate and/or nitrate. Natural acidification processes do occur but their effects appear greatest in coloured surface waters. Land use changes, such as fires, logging, and housing developments, have taken place in many areas with low alkalinity surface waters. However, the symptoms of acidification have not been observed in clearwater lakes and streams except in areas receiving high levels of acidic deposition.

Lakes in the Adirondack Mountain range exhibit some of the lowest alkalinity values found in the eastern United States and are located in a zone presently receiving high acidic deposition (30-40 kg/ha.yr of sulphate in precipitation). In this area, 52% of the 214 high elevation lakes sampled in 1975 had pH values less than 5.0. Seven percent had pH values between 5.0 and 6.0. The New York Department of Environmental Conservation has concluded that at least 180 former brook trout ponds are acidic and no longer support brook trout. The factors causing these population extinctions have not been demonstrated.

New England currently receives wet sulphate deposition loadings of 17-40 kg/ha.yr. A study of 95 relatively small low alkalinity lakes in New England for which historical data were available showed that 64% had decreased in pH. However, accompanying historical deposition data are not available. A comparison of present alkalinity values with historical values for 56 lakes indicated that 70% had decreased in alkalinity. Two other studies have indicated pH declines in some lakes surveyed in Maine. The relative contributions of natural and anthropogenic sources to acidification of these lakes is not known.

Data from intensive studies of 17 lakes in the Muskoka-Haliburton area of Ontario currently receiving about 20-30 kg/ha.yr sulphate in precipitation have shown that some lakes with summer alkalinity values up to about 40 $\mu\text{eq/L}$ experience pH depressions to values below 5.5 during snowmelt. One inlet stream was observed to have pH values as low as 4.1 during spring snowmelt. Other inlet streams had pH depressions but pH did not drop as low. Of 2,624 lakes surveyed in Ontario, 50% had alkalinity of less than 200 $\mu\text{eq/L}$, a value that may be regarded as the upper limit for potential effects of acidic deposition; 13% of the lakes sampled in the province had alkalinities below 40 $\mu\text{eq/L}$. While these lakes may be representative of the areas sampled, they may not be representative of lakes located elsewhere in the Shield. In another survey of 199 lakes of the Precambrian Shield of Quebec 7.5% had alkalinity of approximately 50 $\mu\text{eq/L}$ or less. There are about 1.5 million lakes on the Precambrian Shield in the provinces of Ontario and Quebec; but it is not possible at present to extrapolate results of the surveys to the total population of lakes.

Observed changes in aquatic life have both been correlated with measured changes in the pH of water and inferred by comparisons of waters of different pH values. Differences have been documented in species composition and dominance and size of plankton communities in lakes of varying pH. Study results show that the number of species is lower in low pH lakes compared to lakes of higher pH. These differences may have important implications for organisms higher in the food chain, but studies to date have not been done that might establish this connection.

Many species of frogs, toads and salamanders breed in temporary pools formed by the mixture of spring rains and snowmelt and subject to pH depression. Embryonic deformities and mortalities in the yellow spotted salamander, which breeds in temporary meltwater pools, have been observed in New York State where the acidity of the meltwater pools was 1.5 pH units lower than that of nearby permanent ponds. Population densities of the bullfrog and woodfrog were lower in acidic streams and ponds than in those of higher pH sampled in Ontario. These data are very limited and therefore the extent of the problem is unknown.

Atlantic salmon populations have disappeared from nine rivers in Nova Scotia but remain in rivers in the same area having higher pH due to greater alkalinity. Decreases in alkalinity and the pH of water over time have been observed in some low pH rivers in Nova Scotia. However, historical chemical data do not exist for the period of major decline in angling success nor do they exist for rivers in which fish declined.

Detailed studies of watersheds and clusters of lakes have been carried out in regions of North America and Scandinavia containing low alkalinity lakes and streams under a range of sulphate deposition

rates. The results of those studies conducted in North America are summarized below.

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.

Evidence of chemical change exists for some waters in regions currently estimated or measured to be receiving between about 20-30 kg/ha.yr sulphate in precipitation. In Nova Scotia rivers, 40 years of historical records document reductions in angling success for Atlantic salmon in nine rivers of low pH. Records over later periods for other nearby rivers document decreases in alkalinity and pH. In Maine there is evidence of pH declines over time and loss of alkalinity from some surface waters. In Muskoka-Haliburton historical evidence documents loss of alkalinity for one lake and pH depressions in a number of lakes and streams. Fish confined to the inlet of one lake died during spring melt. In the Algoma region there are elevated sulphate and aluminum levels in some headwater lakes.

Long-term chemical 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. In Quebec, sulphate concentrations in surface waters decrease towards the east and north in parallel with the deposition pattern of sulphate. Sulphate concentrations are equal to or greater than the bicarbonate concentration in some lakes in the southwest part of the province. In the Adirondack Mountains of New York comparison of data from the 1930s with recent surveys has shown that more lakes are now in low pH categories. The relative contribution of natural and anthropogenic sources to acidification of these lakes is not known. The New York Department of Environmental Conservation has concluded that at least 180 former brook trout ponds are acidic and no longer support brook trout, although a direct association with acidic deposition has not been established. In the Hubbard Brook study area in New Hampshire there are pH depressions in some streams during snowmelt of 1 to 2 units.

In the watershed studies summarized above, sulphate in precipitation was used as a surrogate for total acid loading. Sulphate in precipitation can be reliably measured. It is recognized that dry deposition of sulphate and sulphur dioxide, and the wet and dry deposition of nitrogen oxides, nitric acid, particulate nitrate and ammonia, as well as other compounds, also contribute to acidic deposition. The use of a single substance as a surrogate for acidic loadings adds unknown error owing to site-to-site variability in: (1) composition of deposition, and (2) ability of watersheds to neutralize incoming acidity. Wet and dry deposition of sulphur compounds appeared to predominate in long-term acidification.

Insufficient data are available to related nitrate deposition to short-term water quality effects. Therefore, we are unable to develop nitrate loading/response relationships.

The terrestrial mapping analysis for eastern Canada has demonstrated that the watersheds in which some surface waters have been observed to experience effects are representative, in terms of soil and geological characteristics, of larger areas of eastern Canada. The level of variability within terrain classes is not known.

An alkalinity map of the U.S. shows the location of regions where the mean alkalinity of most of the sampled surface waters is less than 200 $\mu\text{eg/L}$. There is reason to believe that some of these low alkalinity surface waters could experience effects similar to those noted in detailed study sites receiving similar total acidic deposition loadings. However, quantification of the number of lakes and rivers in both countries susceptible to acidification at specific loading rates would require validation of mapping methodologies and increased information on loading rates and the chemistry of lakes and streams. The present empirical evidence covers a broad spectrum of physical and climatological conditions across northeastern North America and therefore provides a basis on which to make only qualitative judgements regarding relationships between acidic loading rates and effects.

Based on the results of the empirical studies, interpretation of long-term water quality data and studies of sediment cores that have been reviewed, we conclude that acidic deposition has caused long- and short-term acidification of some low alkalinity surface waters in Canada and the U.S. Based on our understanding of the acidification process the Work Group concludes that reductions from present levels of total sulphur deposition would reduce further chemical and biological alterations to low alkalinity surface waters currently experiencing effects and would lead to eventual recovery of those waters that have been altered by deposition.

The U.S. members conclude that reductions in pH, loss of alkalinity, and associated biological changes have occurred in areas receiving acidic deposition, but cause and effects relationships have often not been clearly established. The relative contributions of acidic inputs from the atmosphere, land use changes, and natural terrestrial processes are not known. The key terrestrial processes which provide acidity to the aquatic systems and/or ameliorate atmospheric acidic inputs are neither known or quantified. The key chemical and biological processes which interact in aquatic ecosystems to determine the chemical environment are not known or quantified. Based on this status of the scientific knowledge, the U.S. Work Group concludes that it is not now possible to derive quantitative loading/effects relationships.

1.3 TERRESTRIAL ECOSYSTEM IMPACTS

The effects of transboundary air pollution on terrestrial ecosystems have been reviewed on the basis of direct effects on vegetation, effects on soils, and effects on wildlife.

1.3.1 Effects on Vegetation

Three main pollutants are of concern with regard to vegetation effects. These pollutants are sulphur dioxide, ozone, and acidic deposition. Ozone and acidic deposition occur at concentrations above background levels at long distances from emission sources. Sulphur dioxide is more of a concern to vegetation in proximity to point sources of emissions than at long distances, where dispersion effects can reduce atmospheric levels to those of background.

1.3.1.1 Sulphur Dioxide

Near point sources, the adverse effects of sulphur dioxide on vegetation can be both visible and subtle (without development of visible foliar injury). Visible effects can be associated with both doses of high concentrations of sulphur dioxide over short periods of time and low concentrations over extended periods. However, in a few specific cases, atmospheric sulphur dioxide deposition may have beneficial effects on agricultural vegetation grown on borderline or sulphur deficient soils.

Visible effects of sulphur dioxide have occurred on pine forests in Canada subjected to average growing season concentrations of sulphur dioxide of 0.017 ppm. Visible injury to the perennial foliage of coniferous trees results in premature needle drop, reduced radial and volume growth and early death of trees. Reduced growth and yield of crops without the development of visible injury have also been found in certain field experiments.

Annual doses of sulphur dioxide of 0.02 ppm have been associated with habitat modifications in grasslands and the elimination of certain sensitive species of lichens near point sources. Lichens may be markedly affected by sulphur dioxide and are considered as bioaccumulators of very low level sulphur dioxide exposures. Direct effects including visible injury, effects on reproductive capacity and species mortality have been encountered in the field at concentrations of sulphur dioxide as low as 0.006 - 0.03 ppm annual average.

Despite such documented evidence of instances of direct effects, obviously not all, but probably most exposures to sulphur dioxide on a regional scale are below levels producing phytotoxic reactions. However, long-term, low-dose studies have demonstrated direct effects on lichen communities and indirect effects on several plant species.

1.3.1.2 Ozone

Ozone is the most important long-range transported pollutant with respect to vegetation effects. Air masses carry ozone and its precursors over long distances and can affect crops and forests in rural areas remote from sources. As a specific example, ozone related crop injuries in southern Ontario have been reported associated with high ozone levels in air masses moving across Lake Erie. In the U.S., experimentally derived crop yield losses ranging from 2 to 56% (crop dependent) were equated with seasonal 7 hr/day mean ozone concentrations of 0.06 - 0.07 ppm. Yield losses in the various crops were as follows: kidney bean 2%, soybean 10%, peanut 14-17%, and lettuce 53-56%. Although direct effects of ozone have been documented on forest growth, an estimate of loss is difficult to calculate because of the limitations stated in the main report.

1.3.1.3 Acidic Deposition

Acidic deposition in the form of simulated rain has been demonstrated to induce a variety of direct and indirect effects on plants grown under greenhouse or semicontrolled conditions. Foliar injury, growth reductions, and growth stimulations have been found under these growing conditions following treatment with simulated acidic precipitation. However, visible foliar injury has not been documented in the field for vegetation exposed to ambient levels of acidic precipitation. The potential effects of acidic deposition on forest growth have been difficult to assess because of the complicating influence of other environmental and climatic factors. To date, there have been too few studies to establish a clear relationship on the interactions of acidic deposition/sulphur dioxide/ozone to reach a definitive conclusion on effects.

1.3.2 Effects on Terrestrial Wildlife

Direct effects of acidic deposition on terrestrial wildlife have not been reported and are not considered likely. Nevertheless, in some instances, indirect effects have been suggested through three possible mechanisms:

- 1) contamination by heavy metals mobilized by acidity;
- 2) reduction in nutritional value of browse or food source;
and
- 3) loss of browse species or impairment of habitats.

1.3.3 Effects on Soil

Soils vary widely with respect to their properties, support different vegetation communities, are subjected to different cultural

practices, are situated in different climatic zones, and are exposed to a broad spectrum of acid loadings. The following effects of acidic deposition probably occur and in some cases are supported by observation, although the number of field situations where investigators have been able to attribute acidity to precipitation or to compare present with former soil pH value is small.

On soils derived from calcareous parent materials, the effects of acidic deposition will lead to only insignificant increases in lime requirement, except in situations near strong point emitters. Heavy metal deposition from these same point source emitters may also cause soil toxicities.

On acid soils, the absence of clear effects upon tree growth from radial-increment measurements covering several decades suggests there will be no short-term effects attributable to acidic deposition.

From the few field situations where earlier investigations permit a comparison over a reasonable time-frame, there is evidence that less acutely acid soils increase in acidity and lose bases at a faster than normal weathering rate. For acutely acid soils, pH may show only minor changes, while over the same period moderate to appreciably larger amounts of soil aluminum are mobilized. These depend upon whether the forest cover is deciduous (e.g., beech) or coniferous (e.g., spruce).

From one comprehensive field investigation, it has been suggested that the additional amounts of aluminum brought into solution kill feeding roots and permit the invasion of fungi causing tree "dieback", but it is not known whether this phenomenon would occur on other sites and soils. What appears well established from a variety of hydrological, limnological and catchment studies is that acidic deposition can lead to the release of additional amounts of soluble aluminum, thus disturbing previous aluminum/calcium ratios in soils, sediments and streamwaters. An eventual reduction in base status and fertility is suggested.

The sulphate component of acidic deposition appears to be adsorbed by soils containing active aluminum and iron oxides, but where these are absent or present in limited amounts, sulphate functions as a balancing anion, leading to the leaching loss of bases and other cations.

The fate of the nitrate component depends upon wet precipitation/snowmelt characteristics. Nitrate, reaching the surface organic horizons of acid forest soils is held there for assimilation by tree roots during the growing season. There are, however, forested catchments in the northeast where nitrate is passed to water bodies.

The lack of appropriate experimental approaches from which the effects of acidic deposition on soil might be assessed and safe deposition ceilings estimated, has caused scientists to exploit

indirect or special situations. These include working near strong point sources, studying soils treated with acidifying fertilizers, and designing lysimetric experiments incorporating simulated acid rains. From such approaches, a variety of soil effects have been demonstrated, usually of an undesirable nature, but at the present time the problem remains of quantifying the dose-response reactions in the field situations.

1.3.4 Sensitivity Assessment

Regions which may be sensitive to acidic deposition have one or more components (i.e., forests, aquatic life, soil, or water) susceptible to degradation under the influence of acidic deposition. Relative sensitivity of these components is reflected in the rate at which an ecosystem component degrades under a particular acidic deposition loading. Different underlying criteria have to be used to represent sensitivity for the different ecosystem components, such as rate of tree growth, characterization of the soil-base status, or water alkalinity. Because so little is known about the acidic deposition dose-response relationships, the underlying criteria are often imprecise. Therefore, relative sensitivity can only be approximately represented or mapped, and then perhaps for only a few species, ecosystems or theoretical effects.

Attention is focused on the sensitivity of soils and bedrock because results from studies which address vegetation and ecosystem effects are limited and not well understood at this time. In the approach used, the emphasis has been to map a combination of potentially important soil attributes as a best available indicator of relative sensitivity. Soil attributes incorporated include texture, depth to carbonate, pH and cation exchange capacity, as well as glacial and bedrock features. Incompleteness of survey data for certain important properties (e.g., sulphate adsorption capacity, internal proton production, and the role of dry deposition) precludes their use in identifying detailed sensitivities of land or aquatic resources. As far as possible, the eastern parts of the United States and Canada are mapped using a similar conceptual framework which indicates the general extent of areas of different possible sensitivities to the effects of acidic deposition. The significance of these categories will increase as more effects are documented.

1.4 HUMAN HEALTH AND VISIBILITY

1.4.1 Health

Available information gives little cause for concern over direct health effects from acidic deposition. The potential indirect health effects associated with transboundary air pollution discussed are:
(1) contamination of the food chain with metallic substances,

especially mercury; (2) leaching of watersheds and corrosion of storage and distribution systems, leading to elevated levels of toxic metals; and (3) health implications of recreational activities in impacted waters.

The principal conclusions of the report are as follows:

Acidification of lakes is a concern because it may be related to increased mercury contamination of the food chain, thus increasing the health risks associated with high levels of consumption of contaminated organisms. A correlation exists between low pH in lakes and higher mercury concentrations in some species of fish, although the mechanism for this accumulation is not presently known. In addition to the effects produced by acidic deposition, the increased input of anthropogenic sources (air or water effluents) of mercury and other heavy metals may further complicate the issue and lead to health problems when affected fish are consumed by humans in large amounts.

Acidic deposition may liberate metals in some groundwaters, surface drinking water supply systems and cisterns. However, groundwater may also be acidic due to increased partial pressure of CO₂ at depths of a few metres or more. This should not be confused with acidity due to atmospheric deposition. Elevated metal concentrations in acidified drinking water supplies have been found. Lead levels in tap water from cisterns were much higher than those found in the source water; about 16% of the households sampled in one western Pennsylvanian county had tap water levels in excess of the United States drinking water standards. Surface drinking water supplies which are not treated (i.e., small communities or individual water supplies) are susceptible. No adverse health effects resulting from consumption of such water have been reported. Concern has been expressed that recreational activities in acidified waters, such as swimming, may prove to cause eye irritation. To date, no compelling evidence has been forthcoming that indicates that humans are being adversely affected by these waters in their current state.

With respect to the direct inhalation of transported air pollutants for which standards exist (i.e., particulate matter, photochemical oxidants, sulphur oxides, and nitrogen oxides), no adverse human health effects are anticipated, providing the ambient air quality standards are not exceeded (see Table 5-2). However, in regions where transboundary air pollution contributes to the violation of the standard, health related problems cannot be ruled out.

Although some concern has been expressed over the effects of acid sulphates on mortality/morbidity, the available data appear insufficient to single out this species as the sole pollutant of

concern in the sulphur-particulate complex. As with the gaseous pollutants, the long-range transport of particulate matter should only be viewed as a concern when violation of the ambient air quality standards occur.

1.4.2 Visibility

Effects of transboundary air pollution on visibility are related to fine particle air quality and only indirectly to acidic deposition. The major precursors of acid deposition that can significantly affect visibility are sulphuric acid and various ammonium sulphate aerosols. These form a large fraction of the fine particle loadings that dominate visibility impairment from anthropogenic sources. Available data do not suggest that nitrates (predominantly in the vapour phase) play a significant role in impairment of visibility, but visible brown plumes from NO_2 have been reported at a distance of 100 km from a few isolated point sources.

From available information on background and incremental fine particle loadings and relative humidity, estimates of visibility impacts (reduction in visual range and contrast, discolouration from haze or plumes) can be made. Analysis of airport data indicate a substantial decline in regional summertime visibility in the eastern U.S. and portions of southern and eastern Canada between 1950 and 1975, with stable or small improving trends since that time. These changes may be associated with changes in the level and distribution patterns of sulphur oxide emissions.

Areas such as those found in western North America, are the most sensitive to visibility degradation. Usually, good visibility is valued most highly in natural settings such as parks and wilderness areas. Any area, however, with normal viewing distances of a mile or more may be affected by episodic regional haze carrying acid precursor substances. Studies of the value of visibility and public perception indicate that the public cares about visibility and is willing to pay for maintaining or improving it. Accurate economic assessments are not, however, available for eastern North America.

1.5 MAN-MADE STRUCTURES

Certain airborne chemicals can accelerate deterioration of materials. There is evidence that materials in urban areas of Europe and North America have suffered and are suffering from exposure to these pollutants. Materials at risk include statuary and structures of cultural value as well as commonly used construction materials. In the present discussion, exterior surfaces are the focus of interest.

It is reasonable to assume that acidic deposition due to long-range transport and transformation of air pollutants contributes somewhat to material effects. Current understanding of material decay

processes leads to the tentative conclusion that local sources of corrosive pollution mask the effects resulting from long-range transport of acidic deposition.

The principal findings of the Work Group are:

The majority of sensitive materials tend to be located in urban/suburban areas. However, materials at risk cannot be assumed to be proportional to population density.

Relationships between concentration of corrosive gases and damage are better documented than relationships between acidic precipitation or particulates and deterioration.

The main groups of materials which are damaged by outdoor air pollutants are: metals, coatings and masonry. The pollutants are delivered to the surfaces in wet and dry form.

It is generally accepted that SO_2 is the primary species causing damage to materials. The importance of nitrogen compounds is closely related to its particular species and may increase with the predicted increases in NO_x emissions relative to SO_2 emissions.

Chemical degradation processes include deterioration of calcareous building materials by the removal of calcium carbonate through conversion to calcium sulphate and the removal of protective corrosion products on metals, particularly zinc and copper.

Mechanical deterioration of masonry occurs when calcium sulphate enters the porous material and causes internal rupturing due to the pressure of crystallization or hydration.

Regional field studies, chamber tests and atmospheric corrosion sites have indicated the nature and extent of accelerated corrosion associated with metal-pollutant interactions. Dose-response relations have been determined for SO_2 and low-carbon steel and zinc. In some areas of eastern North America, urban centres have experienced extensive and significant deterioration of zinc coverings.

Common materials of construction at risk include, limestone, carbon steel and galvanized steel sheet. Carbon steels must be coated in order to provide useful service life and, thus the coating becomes the material at risk.

Dose-response relations have been determined for sulphur dioxide and ozone for some paints and coatings. In some urban centres, ozone can have a significant impact on the durability of elastomers.

For porous materials such as masonry, the long-term accumulation of pollutants is a major concern especially for deterioration associated with sulphate.

Materials at risk and some active corrosion agents have been identified in numerous field and laboratory tests. Confidence in dose-response relationships is weakened in some cases because of incomplete monitoring of air quality and meteorological parameters in field tests.

1.6 METHODOLOGIES FOR ESTIMATING ECONOMIC BENEFITS OF CONTROL

Traditionally, the decision-making process has required an appreciation of the costs and benefits associated with following a prescribed set of actions. Basic to this process has been the transformation of the implications of these actions, (i.e., converting changes in crop yield and fish catches, into comparable units of measurement). Monetary units are widely accepted as providing comparable weighting units for individual variables. In order to provide the Canada/United States Coordinating Committee with guidance in this important area, the Work Group has undertaken a review of the methodologies available for assessing the economic benefits of controlling long-range transport of air pollution.

The following are the conclusions of the Work Group:

A number of methodologies have been reviewed but presently the basic conclusion of this effort is that application of available approaches for conducting a benefit/cost analysis must either omit real but intangible benefits or include a wide uncertainty range. Despite these real limitations, these methodologies can provide a useful estimate of benefits for some sectors.

There are several techniques which can be applied to determine the primary economic benefits associated with a particular receptor category recognizing that option and legacy values are not captured. However, the lack of data on dose-response relationships limits the application of most of these techniques at this time. For some sectors, differences in producers' income may provide benefit estimates even in the absence of explicit dose-response data.

The value of the secondary benefits can be estimated for specific economic sectors and regions, to derive a partial estimate of the impacts in various geographical areas.

It is evident that more economic research is required. Economic techniques have yet to be rigorously tested in some sectors, such as historical value, and are limited in their treatment of option and legacy values, and in dealing with the issues of property rights.

The initial design of future research efforts to document the effects of acidic deposition should reflect the data requirements for an economic benefit estimate. Interdisciplinary cooperation at the design stage is the best way to ensure results which are amenable to economic analysis.

1.7 NATURAL AND MATERIAL RESOURCE INVENTORY

1.7.1 Introduction

A natural and material resource inventory is a necessary component of an assessment of the benefits of emission reductions. Consequently, the Work Group attempted to compile an inventory for aquatic, terrestrial and man-made resources.

In all cases, the sectoral inventories are incomplete and sometimes lacking in sufficient detail. For example, not only does the aquatic inventory not include an accurate accounting of lakes and streams with their associated alkalinity, but it also does not include a consideration of the population size and diversity of aquatic organisms depending on the maintenance of a stable aquatic environment. Similarly the terrestrial inventory has been limited to only a consideration of hardwoods and softwoods because a comprehensive inventory at the species level is presently lacking.

The inventory has been established on the basis of sulphate deposition regimes coincident with the location of terrestrial features such as soils and bedrock which have a limited capacity to reduce the impact of acidic deposition on aquatic regimes. In no cases were there sufficient data to indicate which particular resources are being damaged by acidic deposition. Thus, this inventory is a categorization of resources potentially at risk, rather than a list of resources now adversely affected by acidic deposition. The completion of this inventory has served to underline the considerable weakness which exists in our ability to adequately quantify the extent of the resource at risk.

1.7.2 Aquatic - United States

Approximately 36,000 km² of the eastern U.S. surface water area (25%) is located in areas of low and moderate potential to reduce acidity (high and moderate sensitivity) and of deposition greater than 20 kg/ha.yr sulphate in precipitation. Only 24% are located in areas with a high potential to reduce acidity (low sensitivity) and of deposition greater than 20 kg/ha.yr sulphate in precipitation. The actual surface water area would be more restricted if data had been available on surface water chemistry (i.e., alkalinity). Additional refinements on the inventory should include data on this

variable as well as more accurate measurements of surface water area.

1.7.3 Aquatic - Canada

Approximately 52,000 km² of surface water area is estimated to be at risk in areas with deposition exceeding 20 kg/ha.yr. Of this total, about 54% (28,000 km²) is located in areas with a low potential to reduce acidity (high sensitivity). The inventory could be improved by better data availability on actual surface areas of waters and kilometres of rivers and streams. Moreover, actual data on aquatic alkalinity and aquatic biota will be required to define more accurately the extent of the resource at risk.

1.7.4 Agriculture - United States

Major crops in the eastern U.S. (corn, soybeans, hay, wheat, tobacco and potatoes) are grown under varying sulphate deposition regimes. Soybeans and tobacco are the only ones, however, with approximately 20% of their yield grown under sulphate deposition greater than 40 kg/ha.yr. For the other crops, less than 10% of their total yield is grown under sulphate deposition greater than 40 kg/ha.yr.

1.7.5 Agriculture - Canada

Many of Canada's most valuable crops are grown in areas of high deposition. These include both grains and vegetables. Importantly, for 6 of the 12 crop types included in the inventory, more than 50% of their individual total yields is grown in areas where sulphate deposition exceeds 40 kg/ha.yr. Only 4% or less of each crop is grown in areas experiencing annual deposition levels of 10-20 kg/ha.yr sulphate in precipitation.

1.7.6 Forests - United States

The annual forest growth in those states east of the 100° meridian in 1977 was 476 million m³. Approximately 10% of this combined hardwood and softwood growth occurs under sulphate deposition regimes greater than 40 kg/ha.yr. Over 75% of the growth occurs under sulphate deposition regimes between 20-40 kg/ha.yr.

1.7.7 Forests - Canada

Canadian forest growth occurs in a slightly different pattern than in the U.S. Of the total annual yield of 150 million m³, about 10% of the hardwood growth is located in the highest deposition area, but only 1% of the softwood growth and 8% of the mixed growth.

Approximately 64% of the hardwood and 70% of the mixed growth occurs in the area of moderate deposition, but only 28% of the softwood growth.

1.7.8 Man-Made Materials - United States

There is no adequate U.S. inventory of renewable or cultural resources. Past efforts to create an inventory of renewable resources have combined per capita material estimates and census data on population distribution. These per capita estimates have been shown to be very site specific and are not an adequate basis for creating a national inventory. The only inventory prepared by the Work Group is one on historic resources exposed to various levels of ambient sulphur dioxide.

1.7.9 Man-Made Materials - Canada

As in the case in the U.S., Canada has no adequate inventory of renewable materials or cultural resources. The historic resources inventory includes historical landmarks, buildings and monuments and parks. The inventory presented here indicates the numbers of each of these which are located in 2 categories of deposition: greater than 40 kg/ha.yr and under 40 kg/ha.yr. Geographically, these resources are located in the area around Quebec City, one of the earliest towns in Canada, and in southwestern Ontario (Windsor-Sarnia).

1.8 LIMING

Mitigation of the effects of acidic deposition by adding neutralizing agents to the receptors has been an obvious action to be considered. Limestone is most often used although other chemicals have been tried. The term "liming" has often been used to describe such treatments and in this section will be used to describe artificial neutralization experiments regardless of the chemical or chemicals actually used.

Extensive work has been carried out on aquatic systems affected by acidic deposition. However, the application of lime products to aquatic resources will not address the potential for damage to forests or buildings and structures.

1.8.1 Aquatic Systems

Liming will not eliminate all problems associated with acidification of surface waters but may be necessary on a limited basis as a means of temporarily mitigating the loss of important aquatic ecosystem components. However, it cannot be used in all situations.

Further, its long-term viability and impact on fish populations needs additional study.

The following observations support this overall conclusion:

Liming can only treat certain aquatic situations, mostly lakes, and must be repeated periodically. It is not practical to locate and treat small temporary meltwater pools because of their large number and widespread occurrences. These pools, however, are an important habitat for amphibians and dependent wildlife. The technology for reliably treating high discharge rivers (such as the salmon rivers of the eastern North American coast) is not available.

Swedish experimental liming programs report some success in being able to promote the growth and reproduction of fish populations. However, all results to date are from experiments which have been run for five years or less. The long-term effectiveness of liming to protect aquatic ecosystems is not known. As a result of liming acidic waters, aluminum poisoning of salmon and rainbow trout has been encountered.

No experimental data on liming are available for surface waters containing some of the important sport fish species in North America, such as muskellunge, walleye and bass.

Anthropogenic acidic deposition will alter the original uniqueness of "wilderness" aquatic environments. The additions of neutralizing agents will further modify the character of these ecosystems and will not preserve the "wilderness" nature of these waters.

1.8.2 Terrestrial Liming

The liming of forest lands to neutralize potential acidic deposition effects on terrestrial ecosystems has serious limitations. These include evidence that liming would not prevent direct foliar injury; that under certain conditions lime additions can disrupt important soil biological relationships and adversely affect forests; and that the area coverage required would tend to be so large as to be economically prohibitive.

1.8.3 Drinking Water Supply

Liming techniques have been effectively applied to the treatment of low pH municipal supplies. The per capita costs range from \$0.18 to \$0.57.

EXECUTIVE SUMMARY

WORK GROUP 2

Atmospheric Sciences and Analysis Work Group

Final Report

Executive Summary

As specified by the U.S./Canada Memorandum of Intent (MOI) of 1980, the major objective of the Atmospheric Sciences and Analysis Work Group (Work Group 2) has been to provide technical advice for the development of a bilateral agreement on transboundary air pollution. Specifically, the Work Group has developed information on the application of current long range transport models and acidic deposition monitoring data for this purpose.

This Final Report is a summary of more detailed technical information provided in four sub-group reports to be published separately. The report discusses these technical matters using terms defined in a glossary.

Acid rain occurs in eastern North America within and downwind of the major source regions of oxides of sulfur and nitrogen. This geographical association between the region of the largest North American emissions of sulfur and nitrogen oxides and the region of the largest wet deposition of sulfur and nitrogen acids constitutes the strongest evidence of an anthropogenic origin for much, if not most, of the acidic deposition in the northeastern U.S. and eastern Canada. Furthermore, there is no doubt that polluted air can readily cross the Canada-United States border in either direction.

The modeling effort by Work Group 2 has tried to quantify the origin of the sulfur falling on various parts of eastern North America. The adequacy of available models to predict the results of alternative emission patterns (i.e., the change in deposition that would result from a change in emissions) is uncertain.

The modeling effort has been directed toward sulfur and its compounds because relatively more information is available about sulfur oxides than for other chemical species. Further Work Group 2 has focused its efforts on emissions and depositions encompassing large time and space scales over

eastern North America. With regard to other species, atmospheric models have not yet reached a stage of development suitable for application to formulating, or understanding the impact of, control strategies for reducing the long range transport of nitrogen oxides, ozone, toxic organics and heavy metals. Progress in modeling long range transport has been encouraging, in part due to stimulus from the Memorandum of Intent process, but many significant modeling unknowns persist. Vigorous research to overcome these unknowns continues in both countries.

The main conclusions of the Work Group are summarized below:

Observations of Deposition

- o Monitoring data in North America show a strong geographical correspondence between a large contiguous region of precipitation having low pH (that is, below 4.5) and the region of the most intense emissions of sulfur and nitrogen oxides. The region with low pH also corresponds closely with the region having the highest concentrations and depositions of sulfates and nitrates in precipitation and in both cases the maxima are over and immediately downwind of the major source regions. Maps of sensitive ecosystems produced by Work Group 1 make it apparent that this region of high deposition covers extensive sensitive areas in both eastern Canada and the eastern United States. There is another extensive region of low pH precipitation which can also be geographically associated with a major source of anthropogenic emissions; namely, the highly populated, industrial region of Europe.

- o On a global basis, the presence of occasional, very acidic (low pH) rains in extremely remote areas suggest that there are possible natural and/or very distant anthropogenic sources of sulfur and other compounds that are capable of contributing to the acidity of precipitation. The sources of such substances producing acidity in these remote locations has not yet been determined but may be due to the relative absence of buffering substances (such as calcium) and the presence of organic acids

in the precipitation. The role which such natural or very distant anthropogenic sources of acidity play in eastern North America, although likely to be small, remains to be clarified in order to determine what "background" deposition to use in constructing atmospheric models of source-receptor relationships.

- o Monitoring data at individual stations have been classified according to the trajectory that the air mass appeared to have taken during the preceding time period. Observations at monitoring sites, such as those in Ontario, New York State, Illinois and Bermuda generally indicate higher sulfate and nitrate concentrations in the air and in precipitation when the air mass has passed over areas of higher emissions. While such back air trajectory analyses reinforce the conclusion that man-made emissions have a major influence on acidic deposition, this method is unable to distinguish between near and more distant sources within the same directional sector and cannot be used to trace an air mass trajectory during periods of weak, variable air flows or over very long distances.
- o Although the historical data are of poor quality and limited quantity, one may note that in the eastern U.S.: a) the lack of a noticeable trend in wet sulfate deposition during approximately the past 5 years is consistent with the small changes in sulfur dioxide emissions; and b) an upward trend in the wet nitrate deposition agrees with a similar upward trend in nitrogen oxide emissions.
- o Acidic deposition can occur via precipitation (rain, snow, etc.) or through dry deposition involving gaseous or particulate species. Wet deposition measurements are available for a large number of chemical species. Experimental techniques to monitor dry deposition levels are under development and currently available estimates of dry deposition levels have not been verified by direct observations.

Mass Budget Studies and Man-Made versus Natural Sources

- o Gross budget studies over large regions have examined the major input (including natural) and output quantities of sulfur compounds in the atmosphere. Studies for North America indicate that man-made sulfur oxide emissions exceed natural emissions by a factor of 10 to 20. Results derived from simple climatological analyses indicate that about three to five times more sulfur flows north, from the U.S.A. to Canada, than south. The ratio of the USA to the Canadian emissions of sulfur is almost six to one.
- o Budget studies also suggest that in eastern North America the three removal terms, wet deposition, dry deposition and outflow (into the Atlantic), are roughly equal. Further work is underway to determine the validity of this estimated apportionment.
- o Mass budget studies are in progress for nitrogen oxides and other species.

Local Versus Long Range Transport

- o Deposition values at the more remote pristine locations in eastern Canada and in other remote areas worldwide clearly cannot be attributed to local sources, which are negligible, and demonstrate the reality of a long range transport component.
- o While this report concentrates primarily on the long range transport aspects of transboundary pollution, it is recognized that near-source (less than 50 kilometers) and mesoscale (50-300 kilometers) phenomena are important.
- o Areas in eastern North America experiencing high wet deposition are generally situated in the vicinity (less than 300 kilometers) of non-negligible sources of emissions, while, at greater distances, lesser wet deposition rates occur over wider areas. In the context of the acid rain issue both scales must be considered.

- o The proportion of emitted material deposited in the first 50-100 kilometers is highly variable.

Atmospheric Transformations and Depositions of Sulfur Compounds

- o Sulfur compounds can be transported and transformed by a variety of chemical and physical processes. As a result of these processes, a number of different sulfur compounds are deposited. Our understanding of these processes varies depending upon the compound under consideration and the manner in which it is deposited.
- o The transformation of sulfur dioxide to sulfate aerosol and/or sulfuric acid in the atmosphere is a key aspect of the acid deposition problem.
- o Over the shorter time and space scales, all of the important sulfur dioxide chemical conversion processes are non-linear. Current long range transport model studies by the Work Group make linear approximations of these various chemical processes and may be subject to error in the prediction of the depositions of individual sulfur species (e.g., sulfates). Such modeling errors would also apply to predictions of the change in deposition of individual sulfur species that would result from a change in emissions. Because of the lack of scientific information on some of the conversion reactions, an estimate of the magnitude of errors due to non-linear processes is unavailable.
- o Applications of present models to the prediction of total sulfur, rather than individual species, deposition may reduce the importance of non-linear chemistry. Likewise, the effects of non-linear processes may diminish as larger space and time scales are used. The magnitude of the reduction in error, if any, resulting from such aggregation is not known at this time.

Long Range Transport Models as Assessment Tools

- o Concentrations and depositions (both dry and wet) of sulfur compounds are predicted by long range transport models using simplified

formulations. These simplifications result from (1) an incomplete understanding of some of the physical and chemical processes; (2) limitations in the data available for model input; (3) the paucity of data for testing deposition simulations and (4) the difference between the model and process scales.

- o Using available wet deposition measurements, current models are able to reproduce the correct order of magnitude of the large time and space scale features of measured wet sulfur deposition fields.
- o Model evaluation, that is, the statistical comparison between model predictions and observed values, is not yet considered to be complete. For the 1978 data set, most models appear to perform relatively better in predicting the deposition of sulfate in precipitation than in predicting sulfur concentrations in ground level air.
- o The eight long range transport models have been used to produce tables (that is, transfer matrices) which relate sulfur emissions from specific regions to the deposition of sulfate in precipitation and ambient sulfur concentrations in specific regions. The transfer matrices of the different models exhibit variations among the magnitudes of the transfer matrix elements. This variability could lead to substantial differences in the selection of optimum emission reduction scenarios depending upon the particular model applied and the level of detail required. The long range transport models examined by the Work Group predict generally similar relative sulfur depositions on receptor regions in terms of ranked order of importance. It has not been possible to date to choose a "best model" among the eight or to produce with confidence a "best estimate" single transfer matrix for each variable based upon a valid statistical analysis of all model results.

In accordance with its instructions the Work Group has provided, in its final report, recommendations for the research, modeling and monitoring aspects of a transboundary air pollution agreement.

EXECUTIVE SUMMARY

WORK GROUP 3B

A. SUMMARY OF FINDINGS AND CONCLUSIONS

A.1 INTRODUCTION

Reviewed in this report are emissions of pollutants that undergo transboundary transport, the currently available technologies (process and control), costs of their application for the reduction of SO₂ and NO_x emissions for both new and retrofit installations and research and development activities. The cost for control equipment is a function of the degree of control desired, and is greater for retrofit installations than for new installations. While no detailed intersectorial analysis has been carried out for control costs it would appear that on a per unit of reduction basis, SO₂ controls may be far more costly for certain sectors than others. In addition, the impact of imposing controls on industry may not be limited to control costs. The problems of raising large capital sums, the payback period, the effect of tax and incentive legislation and the difficulty of bearing annual costs will all require detailed examination in the development of optimal control strategies.

Emissions (historical, present and projected) are listed for the fossil-fuel-fired electrical generation sector (eastern U.S. and Canada), non-ferrous smelters (eastern Canada) and mobile sources (U.S. and Canada). These sectors, together with industrial, residential and commercial fuel combustion, account for the majority of anthropogenic SO_x and NO_x emissions in the eastern part of North America, and hence are judged to be the most important sources in the acid precipitation problem. A brief review is carried out for petroleum refining, solid waste incineration and the pulp and paper industry. These sectors are considered to be of secondary importance to the acid precipitation problem since their emissions of SO_x and NO_x are considerably smaller in magnitude than those of the three primary sectors. A preliminary estimate of the emissions inventory for certain other air pollutants including primary sulfates, volatile organic compounds and selected metals is also provided.

To date, regulatory activity has been centered on maintaining and improving local and regional air quality. From the emission projections contained in this report it appears that between 1980 and 2000, SO₂ emissions will increase 7.6%; NO_x emissions will increase 26%. For the eastern portion of the U.S. and Canada (26 states east of the Mississippi River and provinces from Manitoba eastward) SO₂ emissions will decrease by 1% while NO_x emissions will increase by 38%. For certain source sectors and certain emitting regions increases in emissions may be significant.

Research & Development for SO_x and NO_x control for combustion sources is currently centered in three principal areas of activity; improvement in flue gas desulfurization technology, combustion modifications and fluid-bed combustion. It is considered that this research is necessary and should be supported to the maximum practicable extent. Some of these current initiatives

may become commercially viable within the next 10 years; for example, combustion modifications offer the promise of relatively inexpensive NO_x control for coal-fired power plants.

A.2 EMISSIONS SUMMARY

Historical, present and projected emissions of sulfur dioxide and nitrogen oxides, and estimates of the probable error ranges around the present emissions in Canada and the United States have been developed. Emissions projections are based on baseline assumptions about economic and energy growth, assuming no changes in current environmental regulations. In addition, preliminary estimates of emissions of primary sulfates, volatile organic compounds and selected metals have been assembled.

Emissions of SO₂ in the U.S. rose from close to 20 million tonnes in 1950 to about 28 million tonnes in the mid-1960's before dropping to about 24 million tonnes in 1980. The southeastern and midwestern states shared the bulk of this increase. The southeastern states of Alabama, Florida, Georgia, Mississippi, Kentucky, North Carolina, South Carolina and Tennessee exhibited a sharp increase in SO₂ emissions between 1955 and 1978. The data suggest that this increase may be as high as three-fold, i.e. from about 2.1 million tonnes in 1955 to about 5.3 million tonnes in 1978.

In the midwestern states of Illinois, Indiana, Michigan, Minnesota, Ohio and Wisconsin there appears to have been a significant steady increase in SO₂ emissions between 1955 and 1965 (from 6.6 million tonnes to 9.8 million tonnes) and a significant steady decline in these emissions since 1965 to 8.1 million tonnes in 1978. Levels today are about 25% higher than in 1955 in this region of the United States.

Total Canadian emissions of SO₂ were approximately 4.8 million tonnes in 1980, about the same level as in 1955, after having peaked in 1965 at close to 6.6 million tonnes. Eastern Canada, comprising the provinces east of the Manitoba-Saskatchewan border, contributed the bulk of these emissions, i.e. 4.3 million tonnes in 1955, 5.6 million tonnes in 1965 and 4.0 million tonnes in 1980.

Nitrogen oxides emissions in the U.S. increased significantly in all areas over the 1950-78 period. This increase ranged from about a factor of two in the northeast to over three in the south. The trend also indicates that total U.S. NO_x emissions have increased steadily from about 9 million tonnes to 20 million tonnes and did not peak in the mid-1960's as did SO₂. In the eastern U.S., emissions which were at a level of about 6 million tonnes in 1950 reached more than 17 million tonnes in 1978. Total NO_x emissions in Canada have increased from 0.6 million tonnes in 1955 to 1.8 million tonnes in 1980. Eastern Canada has contributed more than 60% to these emissions over this period.

During the 1978-80 period, yearly SO₂ emissions in North America (i.e. both U.S. and Canada) have amounted to close to 29 million tonnes. In eastern North America the total is close to 25 million tonnes, and the ratio of U.S. to Canadian emissions is 5.3 to 1.

Thermal power plants are the primary source of anthropogenic SO₂ emissions and contribute about 57% of the combined U.S.-Canada nation-wide emissions. This sector is followed by the industrial, commercial and residential fuel combustion category at about 14% of the combined nation-wide emissions. Then, at about 12%, are the emissions of SO₂ from non-ferrous smelters with all other industrial processes contributing about 13%. The primary contributor to present domestic SO₂ emissions differs in the U.S. and Canada. In the U.S. about two-thirds of the total domestic emissions comes from power plants, while in Canada more than 40% comes from non-ferrous smelters. About 16 million tonnes of SO₂ come from American power plants, about 2 million tonnes of SO₂ come from Canadian non-ferrous smelters. Only about 15% of the SO₂ generated in Canada comes from thermal power plants.

Yearly NO_x emissions in North America during the same period have amounted to about 21 million tonnes. In eastern North-America the total is about 16 million tonnes, while the ratio of U.S. to Canadian emissions in the east is roughly 15 to 1. About 45% of the combined nation-wide emissions comes from the transportation sector, about one quarter from power plants and about 20% from other combustion processes (industrial, commercial, residential, fuel combustion).

An analysis was performed to estimate the probable error inherent in the current emission estimates of SO₂ and NO_x. The probable errors are approximations derived through a combination of statistical theory and engineering judgement and do not represent true error values obtained through the application of rigorous statistical procedures.

The probable error in the national U.S. SO₂ emissions is estimated to be 2.3% and for NO_x, 2.0%. For Canada, the precision of the national SO₂ inventory was found to be 6.3% and for NO_x, 10.3%. For individual states or small regions, the probable error is higher.

In the next two decades total U.S. SO₂ emissions are projected to decrease slightly by 1990 to about 23 million tonnes from about 24 million tonnes in 1980 and then increase to about 27 million tonnes by the year 2000. Emissions from power plants are projected to remain roughly constant at about 16 million tonnes while emissions from the combustion of fuel in residential, commercial and industrial sectors is projected to increase from 3.2 million tonnes (1980) to 7.4 million tonnes (2000) or roughly 130%. The increase in emissions from the industrial sector is based on the assumption that there will be large increases in coal usage in industrial boilers. Emissions of SO₂ from U.S. non-

ferrous smelters are projected to decrease to 0.5 million tonnes by the year 2000 from 1.4 million tonnes in 1980 as all non-ferrous smelters, due to existing regulatory requirements, must achieve approximately 90% reduction in SO₂ emissions from uncontrolled levels. However, non-ferrous smelters in the U.S are located in western and southwestern states and are therefore unlikely to play a significant role in the eastern North America acid precipitation issue. Emissions of SO₂ from other industrial processes are projected to decrease by about 50% by the year 2000 to about 1.5 million tonnes.

In Canada, total emissions of SO₂ to the end of the century are expected to decrease slightly from 4.3 million tonnes (1980) to 4.4 million tonnes (2000). Sulphur dioxide emissions from thermal power plants are expected to decrease slightly from 0.8 million tonnes in 1980 to 0.7 million tonnes by the year 2000. Although the emissions of SO₂ from power plants in Western Canada are projected to increase from about 80 kilotonnes in 1980 to close to 290 kilotonnes by the year 2000, this increase will be offset by decreases in SO₂ emissions in the Maritime Provinces and by decreases in SO₂ emissions from power plants in Ontario. Recent regulatory requirements announced by the Ontario government will limit the emissions of SO₂ from Ontario power plants to 260 kilotonnes/year by 1990 from the current 400 kilotonnes/year.

Sulfur dioxide emissions from industrial, commercial and residential fuel combustion are projected to decrease significantly from 0.8 million tonnes in 1980 to 0.25 million tonnes by the year 2000. The underlying assumption here, as reflected in the Canadian National Energy Plan (NEP), is the conversion to the use of natural gas from petroleum fuels.

Projected SO₂ emissions from Canadian non-ferrous smelting complexes to the year 2000 indicate a maximum of about 2.2 million tonnes per year, essentially the same level as in 1980. The level attained will depend on market fluctuations assuming no technological improvements are made at the various smelters. Emissions from other industrial processes are projected to increase from about 0.9 million tonnes in 1980 to close to 1.1 million tonnes by the year 2000. This increase is due primarily to an increase in SO₂ emissions in Western Canada based on the assumption that projected tar sands and natural gas development projects will proceed as scheduled. Recent events suggest that this assumption may no longer apply.

With respect to the emissions of NO_x to the year 2000, total U.S. emissions are projected to rise from about 19 million tonnes in 1980 to about 24 million tonnes by the year 2000. In Canada, the emissions are also projected to rise from about 1.8 million tonnes in 1980 to about 2.4 million tonnes by the year 2000.

An increase of about 55% in nitrogen oxides emissions is projected by the year 2000 from U.S. power generating stations, while in Canada the level is projected to be essentially the same as in 1980, i.e. about 250 kilotonnes. In the transportation sector, the emissions in the U.S. are expected to rise from about 8.5 million tonnes in 1980 to close to 10 million tonnes by the year 2000. In Canada, in the absence of further control action, NO_x emissions are projected to rise steadily from about 1.1 million tonnes in 1980, to 1.3 million tonnes in 1990 and to 1.7 million tonnes by the end of the century. Nitrogen oxides emissions both in Canada and the U.S. are projected to remain essentially constant from commercial, residential and industrial fuel combustion.

Present and projected SO₂ and NO_x emissions data for the U.S. and Canada are presented in Tables A.2.1 to A.2.4. As mentioned previously, the emission projections shown assume no changes in current environmental regulations.

Preliminary estimates of the emissions of pollutants that may also be important in the long range transport of pollutants, i.e. primary sulphates, volatile organics and selected metals have also been developed. Current or the most recently available emissions data for these pollutants are shown in Tables A.2.5 to A.2.8.

TABLE A.2.1

UNITED STATES

National Current and Projected SO₂ Emissions
Using Combined Models (10⁶ tonnes/year)²

	<u>Current 1980</u>	<u>Projected 1990</u>	<u>Projected 2000</u>
Electric Utilities	15.8	15.9	16.2
Industrial Boilers and Process Heaters	2.4 ¹	3.4	6.5
Non-ferrous Smelters	1.4	0.5	0.5
Residential/Commercial	0.8	1.0	0.9
Other Industrial Processes	2.9 ¹	1.2	1.5
Transportation	<u>0.8</u>	<u>0.8</u>	<u>1.0</u>
TOTAL U.S.	24.1	22.8	26.6

¹ In current 1980 emissions, process heaters are included with other industrial processes and not with industrial boilers.

² Source of Projections: These emissions estimates are based on 1980 trends using the following sources for projections for each sector: Utility-EHPA; Industrial-ICF; Res/Com-SEAS; Smelters-site by site survey; Industrial Processes-SEAS; Transportation-SEAS.

TABLE A.2.2

UNITED STATES

National Current and Projected NO_x Emissions
Using Combined Models (106 tonnes/year)³

	<u>Current 1980</u>	<u>Projected 1990</u>	<u>Projected 2000</u>
Electric Utilities	5.6	7.2	8.7
Industrial Boilers and Process Heaters	3.5 ¹	3.0	4.0
Residential/Commercial	0.7	0.7	0.6
Other Industrial Processes	0.7 ¹	0.8	1.1
Transportation	8.5	7.8	9.7
Miscellaneous	<u>0.3</u>	<u>-- 2</u>	<u>-- 2</u>
TOTAL U.S.	19.3	19.5	24.1

1 In current 1980 emissions, process heaters are included with other industrial processes and not with industrial boilers.

2 Projections of emissions from miscellaneous sources (solid waste disposal, forest fires, etc) were not produced

3 Source of Projections: These emissions estimates are based on 1980 trends using the following sources for projections for each sector: Utility-EHPA; Industrial-ICF; Res/Com-SEAS; Industrial Processes-SEAS; Transportation-Mobil2

TABLE A.2.3

CANADA

National Current and Projected SO₂ Emissions
(10⁶ tonnes/year)

	<u>Current 1980</u>	<u>Projected 1990</u>	<u>Projected 2000</u>
Electric Utilities	0.75	0.65	0.66
Industrial Fuel Combustion	0.62	0.33	0.23
Residential/Commercial Fuel Combustion	0.21	0.08	0.03
Non-ferrous Smelters	2.13	2.32	2.32
Other Industrial Processes ¹	0.92	1.16	1.11
Transportation	<u>0.16</u>	<u>0.16</u>	<u>0.16</u>
TOTAL	4.77	4.70	4.51

NOTE: Total may not add up due to rounding.

¹ Includes process emissions from petroleum refining, natural gas processing, tar sands operations, other various industrial processes.

² Sources of projections: The projected emission estimates are based on the current 1980 emissions and were developed by Environment Canada using energy consumption projections provided in National Energy Board reports.

TABLE A.2.4

CANADA

National Current and Projected NO_x Emissions
(10⁶ tonnes/year)

	<u>Current 1980</u>	<u>Projected 1990</u>	<u>Projected 2000</u>
Electric Utilities	0.25	0.19	0.26
Industrial Fuel Combustion	0.30	0.30	0.33
Residential/Commercial Fuel Combustion	0.09	0.07	0.07
Non-ferrous Smelters	0.01	0.01	0.01
Other Industrial Processes ¹	0.07	0.07	0.07
Transportation	<u>1.11</u>	<u>1.34</u>	<u>1.67</u>
TOTAL	1.83	1.98	2.41

¹ Includes process emissions from petroleum refining, natural gas processing, tar sands operations, other industrial processes and miscellaneous sources.

² Source of Projections: The projected emission estimates are based on the current 1980 emissions and were developed by Environment Canada using energy consumption projections provided in National Energy Board reports.

TABLE A.2.5

National Current Emissions of
Primary Sulfates (Kilotonnes)

	U.S. ¹		Canada ²
	Total Sulfates	Acid Sulfates	Total Sulfates
Electric Utilities	255	197	15.9
Non-utility Fuel Combustion	157	108	67.8
Non-ferrous Smelters	24.6	16	33.5
Transportation	32.4	24	4.1
Other Sources	<u>114</u>	<u>53</u>	<u>70.7</u>
TOTAL	584	398	192.0

TABLE A.2.6

National Current Emissions of
Volatile Organic Compounds (Kilotonnes)

	<u>U.S.¹</u>	<u>Canada²</u>
Transportation	8 032.1	850.1
Stationary Source Fuel Combustion	1 130.0	102.1
Industrial Processes	11 090.1	413.9
Solid Waste Disposal	697.4	31.2
Miscellaneous	<u>2 427.3</u>	<u>567.8</u>
TOTAL	23 376.9	1 974.1

¹ Estimate for 1980

² Estimate for 1978

TABLE A.2.7

UNITED STATES

Selected Metals Emissions, By Category
(tonnes)

Heavy Metals	Industrial Processes	Fuel Combustion	Trans- portation	Incineration	Misc.	TOTAL
Arsenic	4 720	540	NEG	NEG	2 830	8 090
Barium	5 315	810	NEG	NEG	2 705	8 830
Cadmium	202	455	80	48	NEG	785
Chromium	3 225	670	NEG	40	NEG	3 935
Copper	3 623	660	NEG	155	180	4 618
Lead	4 706	835	43 240	3 363	NEG	52 140
Manganese	13 737	2 575	NEG	NEG	NEG	16 312
Mercury	60	98	NEG	8	330	496
Nickel	885	8 545	810	NEG	NEG	10 240
Selenium*	122	211	NEG	NEG	NEG	333
Vanadium	245	14 235	NEG	4	NEG	14 484

* Non-metallic element

TABLE A.2.8

CANADA

Selected Metals Emissions, By Category
(tonnes)

Heavy Metals	Industrial Processes	Fuel Combustion	Transportation	Incineration	Misc.	TOTAL
Arsenic	3 654	34	Neg	1	5	3 694
Barium	102	118	10	16	---	246
Cadmium	416	86	3	3	Neg	508
Chromium	69	54	12	102	7	244
Copper	2 318	91	5	79	302	2 795
Lead	4 726	30	11 658	301	60	16 775
Manganese	5 972	17	Neg	7	14	6 010
Mercury	20	9	---	2	9	40
Nickel	974	596	55	54	90	1 769
Selenium*	130	47	1	1	Neg	179
Vanadium	24	1 848	---	1	1	1 874
Antimony	52	2	3	4	---	61
Beryllium	Neg	7	---	Neg	---	7
Bismuth	61	19	Neg	2	---	82
Cobalt	78	13	1	2	1	95
Tin	43	39	Neg	85	2	169
Zinc	6 341	134	795	124	2	7 396

* Non-metallic element

A.3 FOSSIL-FUEL-FIRED ELECTRICITY GENERATION - CONTROL TECHNOLOGY SUMMARY

In this sector the focus is placed on fossil-fuel-fired electricity generation, the attendant SO_x and NO_x emissions and their control and reduction. The principal reason for this is that these facilities are important contributors to total emissions of these pollutants. While hydroelectric and nuclear facilities are commercially available forms of power production, in the past these options have been selected on the basis of the availability of natural resources, technical, social and economic considerations and not as an alternative control strategy for SO_x and NO_x emissions. Power generation processes such as magnetohydrodynamics, tidal, wind and solar power have also been excluded from consideration in this report as alternative means for SO_x and NO_x control because currently they are only emerging technologies.

SO_x Reduction

Sulfur oxide emissions can be reduced by several methods. These can be grouped generically as follows:

- (1) use of naturally occurring low-sulfur fuel;
- (2) removal of the sulfur before combustion;
- (3) reaction of sulfur with an absorbent during combustion; and
- (4) removal of the sulfur oxides after combustion.

Some processes for SO₂ control are capable of a very high removal efficiency with attendant expense; others cost much less but are limited to a relatively low level of removal efficiency.

For coal combustion, the following rankings are made for process choice at different levels of emission reduction. They are judgmental in nature, being based on a subjective evaluation of factors such as cost, commercial viability, absorption efficiency, and process reliability and may well change when site-specific considerations are taken into account. It is not considered feasible to undertake a more quantitative approach to ranking in this report.

Removal Efficiency level, %

Process Listing

Higher than 90%

1. Limestone scrubbing with additives
2. Lime scrubbing
3. Dual alkali scrubbing
4. Regenerable FGD processes

50-90% (high-sulfur coal)

1. Limestone scrubbing, (with physical coal cleaning where upper limit on SO₂ emissions applies)
2. Chemical coal cleaning^a

Removal Efficiency level, %

Process Listing

50-90% (low-sulfur coal)

3. Low-sulfur fuel substitution
4. Limestone injection with a multistage burner^a

Below 50%

1. Lime spray dryer process
 2. Limestone scrubbing
-
1. Physical coal cleaning (highly variable effectiveness due to variation in coal properties)
 2. Blending with low-sulfur coal

^a Not presently available commercially

Tables A.3.1 and A.3.2 summarize the relative merits and available cost data for sulfur oxide controls on thermal power plants.

Physical coal cleaning costs up to around \$800 per tonne of SO₂ removed for high-sulfur coals. The capital costs of wet flue gas desulfurization (FGD) at new plants range between \$162-326 per kilowatt of installed capacity. Limestone systems tend to be the most economical wet FGD system. FGD processes such as the dual alkali and Wellman-Lord processes, tend to be more expensive than purely non-regenerable processes. The capital costs of dry scrubbers are \$154-200 per kilowatt of installed capacity but the technology is still under development. In general, there is a wide range in the actual installed cost of FGD systems due to the variables that need to be considered on a site-specific basis.

NO_x Reduction

Several approaches can be used for NO_x control depending upon the degree of control required. Combustion modifications are the most cost-effective methods for low levels of control. Flue gas treatment by ammonia injection methods have now achieved operational acceptance on coal-fired plants in Japan and could be considered if a high degree of control is required. A rough ranking of the degree of control is as follows:

Removal efficiency level, %

Process Listing

90% or higher

1. Catalytic reduction with more than normal amount of catalyst, preceded by combustion modifications

Removal efficiency level, %

Process Listing

50-80%

1. As above, with normal amount of catalyst
2. Combustion modifications (all types) followed by non-catalytic reduction (ammonia injection without catalyst)
3. Combustion modifications alone (for lower levels of removal minimize boiler problems)
4. Low-NO_x burners

Below 30%

1. Staged combustion^a
2. Low-NO_x burners^a
3. Flue-gas recirculation (except for coal^a)

^a Used in combination with others, if necessary, to achieve the required reduction level.

The capital costs associated with combustion modification techniques for the control of NO_x emissions from thermal power plants are estimated at:

<u>Techniques</u>	<u>Capital Cost</u>	<u>Lowest Achievable NO_x Emission Level</u>
Low Excess Air	\$0	0.9 lb per 10 ⁶ Btu
Staged Combustion (over-fired air)	\$2-3/kW	0.7 lb per 10 ⁶ Btu
Low-NO _x Burners	\$2-\$10/kW	0.4-0.5 lb per 10 ⁶ Btu

The capital cost estimates for NO_x control vary considerably due to site-specific variables (e.g., boiler type). The uncertainty in the cost data ranges from -10 percent to +30 percent. Furthermore, the cost of flue gas treatment (FGT) processes for NO_x control have not yet been determined.

TABLE A.3.1
Control Technologies for SO₂ Reduction

SYSTEM	PERFORMANCE	APPLICABILITY	UNCERTAINTY	WASTE DISPOSAL	PROBLEMS
Wet F.G.D. Limestone Lime	Acceptable Availability (90% or less)	All fuels	Cost is a function of size, sulfur content, location, redundancy of equipment, whether ash removal is included. ^a	Preferably oxidized to gypsum, otherwise settling problems in ponds and land- fill unless chemically fixed.	Waste disposal because of volumes. Utilities sceptical of costg and relia- bility. ^a
Dual Alkali	Acceptable Availability	All fuels		As above.	As above.
Wellman Lord	Limited experience so far.	All fuels	Uncertain market by-products.	Potential water pollution problem.	High cost.
Dry Scrubber	Limited experience so far.	Low sulfur fuels	Performance data sparse.	Lime systems have minimal problems, whereas soda-based units have potential water pollution problems.	Waste disposal involves large volumes, Opera- tional difficulties with variations in coal characteristics
Low-Sulphur Fuel		Coal	Incremental costs, availability of supplies.		Boiler derating, effects on precipi- tator, transporta- tion, logistics.
Physical Coal Cleaning	Effective up to 25% sulfur removal.	Used for high pyritic sul- fur coals.	Coal variability and expansion of existing facilities	Water pollution and solid waste disposal.	Energy losses, maintaining quality control.

^a This comment is equally applicable to all processes in this table.

TABLE A.3.2

**Cost of Control Technologies for SO₂ Reduction
for Representative 500 MW Coal-fired Thermal Power Plants**

SO ₂ CONTROL TECHNOLOGY	LOW SULFUR COAL ^b			HIGH SULFUR COAL ^c		
	CAPITAL COST \$/kW	LEVELIZED COST mills/kWh	COST EFFECTIVENESS \$/tonne SO ₂ removed	CAPITAL COST \$/kW	LEVELIZED COST mills/kWh	COST EFFECTIVENESS \$/tonne SO ₂ removed
Wet FGD ^a						
- Limestone	176	10.7	3806	244	16.4	840
- Lime	162	11.3	4026	224	17.4	880
- Dual Alkali	181	11.2	4004	251	17.1	880
- Wellman Lord	235	13.6	4862	326	20.9	1100
Dry FGD ^a						
- Lime	154	9.4	3344	200	17.1	880
- Sodium	158	10.2	3630	-	-	-
Physical Coal Cleaning ^d	-	-	-	-	-	790

a) The source of the costs quoted are reports EPA-600/7-81-014 and EPA-600/9-81-019a. These are hypothetical costs derived from a computer model generated by Tennessee Valley Authority, for a particular set of assumptions, viz. 500 MW unit, located in the U.S. upper mid-western states, burning coal, operating for 5500 hours per year, for 30 years. The capital and operating costs for particulate matter collection are included in the SO₂ reduction costs. It is further assumed that the capital costs are in 1982 dollars, and the revenue is in 1984 dollars. Actual historical cost data are available in EPA-600/7-81-012a Tables A1 and A2 "EPA Utility FGD Survey", Jan. 1981.

b) Low sulfur western coal, 9 700 Btu/lb. 0.7% Sulfur (dry basis); 70% SO₂ removal.

c) High sulfur eastern coal, 11 700 Btu/lb. 3.5% Sulfur (dry basis); 90% SO₂ removal for FGD processes.

d) Source: EPA-600/7-81-086; 28% SO₂ removal.

A.4 NON-FERROUS SMELTERS

In Canada there are a total of five copper smelters, three nickel-copper smelters, two lead smelters and three zinc smelters. The major sources of smelter SO₂ emissions in Canada are copper and nickel smelters located in Manitoba, Ontario and Quebec.

In the United States, there are a total of 15 copper smelters, 5 lead smelters and 5 zinc smelters. The major non-ferrous smelting capacity is located in the Western U.S. with the largest concentration in the Arizona-New Mexico area.

In 1980, SO₂ emissions from non-ferrous smelters contributed about 45% of the total in Canada and 6% in the U.S.

Off-gases from non-ferrous smelters basically fall into two categories, those with strong SO₂ strengths (defined as greater than 4% SO₂) and those of weak strengths (less than 4% SO₂). Strong gas streams can be controlled by using add-on technologies such as acid plants and liquid SO₂ plants. These processes are considered proven and, in most cases, affordable control options. While the treatment of weak gas streams constitutes a more difficult and costly problem, control options are available. These include:

- (1) the use of either regenerative or non-regenerative flue gas desulfurization (FGD) processes;
- (2) the modification of furnaces to produce a strong gas stream through measures such as oxygen enrichment;
- (3) the replacement of sources emitting weak SO₂ streams with alternative modern technology producing strong SO₂ streams, controlled by acid plants.

FGD is practiced by a number of smelters world-wide and each system is unique to its smelter. This is a result of the particular circumstance of each application in terms of the cost for raw materials and the availability of by-product markets rather than technical suitability of the processes.

Upgrading of existing furnace operations to strengthen sulfur dioxide content can be an effective approach to SO₂ control when coupled with FGD systems. Alternative pyrometallurgical processes are of interest because they provide a strong SO₂ gas stream for control by a conventional acid plant, reduction in energy consumption, reduction in gas stream volumes, and reduction in operating costs.

Hydrometallurgical processes eliminate the generation of SO₂ streams. However they are more energy intensive and currently have limited application. A large number of alternative approaches to achieve reductions in SO₂ emissions based upon

various combinations of process and control technologies are technically possible; a number of the more important ones are given in Tables A.4.1 and A.4.2. In considering any approach, it is of paramount importance to consider the unique nature of each smelter. This uniqueness factor is determined by the nature of the ore concentrates and the metallurgy required to successfully treat these concentrates. It is these aspects that govern the selection of a metallurgical process for metal winning and, in turn, the degree of sulfur containment. Each smelter requires an individual technical and economic assessment of feasibility.

TABLE A.4.1
Copper/Nickel Smelter SO₂ Control Systems

Smelter Process				SO ₂ Control System						
Technology	Relative Cost ³	Technology availability	Energy consumption ⁵	Technology	SO ₂ Control %	Estimated Cost ⁶	Technology availability ⁷	Operating reliability	Energy consumption	By-product
Green charge or multi-hearth roaster, reverb., converter	90-110	High	High 106-118	Acid plant on converter	To 50%	52	High ⁴	High	Low	Sulfuric acid
Fluid-bed roaster, reverb., converter (base case)	100	High	High 100	Acid plant on roaster	To 45%	33	High	High	Low	Sulfuric acid
Multi-hearth roaster, reverb., converter	110	High	High 100	Non-regenerative FGD	To 85%	134	Low	Low	High	Sulfur compound for waste disposal
Multi-hearth roaster, reverb., converter	110	High	High 100	Regenerative FGD	To 85%	108	Low	Low	High	Sulfuric acid
Fluid-bed roaster, electric furnace, converter	100	High	Very High 106-156	Acid plant on roaster, electric furnace, converter	To 90%	33	High ⁴	High	Low-Med.	Sulfuric acid
Fluid-bed roaster, reverb., converter	100	High	High 100	Acid plant on roaster and non-regenerative FGD on weak gas streams	90-92%	83	Low	Low	High	Sulfuric acid and sulfur compound for waste disposal
Fluid-bed roaster, reverb., converter	100	High	High 100	Acid plant on roaster and regenerative FGD on weak gas streams and acid plant	90-92%	70	Low	Low	High	Sulfuric acid
Dryer, oxygen-enriched reverb., converter	90	Med.	Med. 90-95	Acid plant	90-94%	52	High ⁴	High	Low-Med.	Sulfuric acid

TABLE A.4.1 (continued)
Copper/Nickel Smelter SO₂ Control Systems

Technology	Smelter Process			SO ₂ Control System						
	Relative Cost ³	Technology availability	Energy consumption ⁵	Technology	SO ₂ Control %	Estimated Cost ⁶	Technology availability ⁷	Operating reliability	Energy consumption	By-product
Fluid-bed roaster electric furnace converter	100	High	Very High 106-156	Acid plant on roaster, electric furnace, converter plus FGD system on weak gas streams	To 95%	44	Med.	Med.	Med.	Sulfuric acid and sulfur compound for waste disposal
Direct furnace smelting, converter (Inco, Outokumpu, Noranda)	80	High	Low 60-80	Acid plant on flash furnace and converter	94-95%	40	High	High ⁴	Low	Sulfuric acid
Direct furnace smelting, converter (Inco, Outokumpu, Noranda)	80	High	Low 60-80	Acid plant on flash furnace plus FGD system on weak gas streams	To 95%	43	Med.	Med.	Med.	Sulfuric acid and sulfur compound for waste disposal
Continuous smelting (Mitsubishi, Noranda)	135	Med. ¹	Low 60-80	Acid plant	98-99%	33	High	High	Low	Sulfuric acid
Hydrometallurgy	135	Low ²	High to Very High 100-200	?	To 99.5%	?	?	?	?	Elemental sulfur

¹ Can be used for clean copper concentrates

² Problems with precious metals recovery, limited operating experience; could be considered for some special cases

³ Capital cost relative to a base case facility of calcine fed reverberatory furnace

⁴ Capture of off-gases from nickel converters and electric furnaces not yet developed

⁵ Smelter energy consumption is relative to base case of calcine fed reverberatory furnace taken as 100%

⁶ Estimated cost per tonne of SO₂ removed in 1980 U.S.\$

⁷ High means technology is used at a number of smelters; medium means technology is used at a few smelters; low means technology is used at only one smelter or is being evaluated at a pilot scale facility

Source: Section C.2. References 1, 3, 4, 15

TABLE A.4.2
Comparison of Cost of Abating SO₂ by Various Options⁽¹⁾

Control Options	Off-gas			Capital Cost 10 ⁶ \$	Operating Cost 10 ⁶ \$	Amortization 10 ⁶ \$	Total Annual Cost 10 ⁶ \$	Cost Per tonne SO ₂ ⁽²⁾ removed \$
	SCFM	% SO ₂	SO ₂ tonnes per day					
<u>Single contact acid plant on strong gas stream⁽³⁾</u>								
- continuous gas only	27 000	12	346	17	1.5	2.5	4.0	33
- variable gas only	49 000	5-8	346	28	2.2	4.1	6.3	52
- continuous gas & variable gas	36 000	6-12	346	22	1.8	3.1	4.9	40
<u>Non-regenerative scrubbing of weak gas stream⁽⁴⁾</u>								
- lime	400 000	1	430	40	17.4	5.7	23.1	154
- limestone	400 000	1	430	47	13.4	6.8	20.2	134
<u>Regenerative scrubbing of weak gas stream⁽⁴⁾</u>								
- MgO and acid plant	400 000	1	430	65	10.9	9.3	20.2	134
- Citrate and acid plant	400 000	1	430	58	7.9	8.3	16.2	108
<u>Replacement of process pro- ducing weak gas stream with modern process⁽⁵⁾</u>								
- existing old smelter process (uncontrolled)				--	26.0 ⁽⁶⁾	---	26.0	---
- modern process	63 000	8	540	215	22.0 ⁽⁶⁾	30.9	52.9	115 ⁽⁷⁾

- All costs are in 1981 U.S. dollars. Capital costs is amortized over 12.5 years at 10% interest.
- Production is based on 350 operating days per year and assuming 100% control of SO₂.
- Figures are derived from "A Study of Sulfur Containment Technology in the Non-ferrous Metallurgical Industry", Economic and Technical Review Report EPS-3-AP-79-8, Air Pollution Control Directorate, Environment Canada, April 1980.
- Non-regenerative and regenerative scrubbing of weak gas cost estimates are taken from the EPA data presented in the Appendices. The cost estimates for the regenerative scrubbing of weak gas stream also include a capital cost of \$20 million and an annual operating cost of \$6.8 million for a 660 tonnes per day sulfuric acid plant.
- The cost estimates are for a copper smelter producing 100 000 tonnes per annum of copper. Modern processes are processes such as the INCO, Noranda and Mitsubishi processes.
- The annual operating cost of a fully depreciated, existing reverberatory based smelter with no SO₂ controls is estimated to be about \$26 million. The operating cost of the modern processes is approximated at \$22 million.
- The difference between the annual operating cost for the facility and the old facility are allocated to SO₂ control. The cost per tonne SO₂ fixed is computed assuming 100% SO₂ capture.

A.5 MOBILE SOURCES SUMMARY

The mobile sources sector is a major source of NO_x emissions (currently 44% of NO_x in the U.S. and 60% in Canada) but SO_x emissions from this sector are negligible. Control technology is available for NO_x and stringent emission limits for the design performance of new vehicles are in place in the U.S. and are being considered in Canada for 1985.

The technology for meeting the current automobile emission standards in the U.S. employs the "three-way" catalyst technology (called three-way because it controls HC, CO and NO_x), coupled with a series of electronic and vacuum sensing devices which detect and control selected engine operating parameters.

Between 1980 and 2000, NO_x emissions from this sector are projected to increase by about 15% in the U.S. and by 50% in Canada. For practical purposes this disparity is accounted for by differences in the light duty, gasoline powered vehicles partly because more stringent emission standards were introduced in 1981 in the U.S. and partly because a much faster growth rate for vehicle miles travelled (VMT) by light duty vehicles is being projected in Canada.

In the absence of changes to federal emission limits in either country, the contribution of the transportation sector to NO_x emissions in the year 2000 is projected to be 40% in the U.S. and 70% in Canada. The opposite trends in the two countries (see 1980 percentages above) is explained primarily by the factors cited above but also by the opposite directions of the trend in electric utility emissions (29% to 36% in the U.S. and 13% to 10% in Canada) and the less stringent emission standards for light duty vehicles in Canada.

A.6 INDUSTRIAL, COMMERCIAL, AND RESIDENTIAL FUEL COMBUSTION

Industrial, commercial and residential fuel combustion accounts for approximately 14 percent of combined U.S.-Canada SO₂ emissions and approximately 20 percent of their combined NO_x emissions. This sector is characterized by a wide diversity in boiler sizes (i.e., from 10⁵ Btu/hr to greater than 250 x 10⁶ Btu/hr), combustion systems, and fuel characteristics. In addition, the technical expertise of the owner/operator varies from the homeowner to the skilled technician. Industrial boilers are the major emitters in this sector.

There are various control options that can be considered to reduce sulfur dioxide emissions from this sector. These are:

- (1) replace sulfur containing fuels such as coal and petroleum with natural gas or electrical energy, or substitute lower sulfur fuel of the same type;
- (2) desulfurize fuels such as heating oils and heavy fuel oils;
- (3) use flue-gas desulfurization techniques to remove sulfur dioxide from combustion flue gases.

FGD can lower sulfur oxide emissions by up to 90 percent. Fluid-bed combustion can achieve a 70-85 percent SO₂ reduction at costs which are estimated to be competitive with FGD. The dual-alkali FGD process is the dominant sulfur oxide control technology for industrial boilers. Sodium-based once-through systems are used in industries which produce a sodium-containing waste stream such as pulp and paper and textile mills (from de-ionizer recharging). There are two installations of the lime-spray-dryer SO₂ control process on commercial boilers in the United States.

As in utility boilers, combustion modification is the principal method of controlling NO_x emissions. In California, several thermal-NO_x (non-catalytic NO_x control) installations have been purchased; however, none is in commercial operation at this time. The NO_x emission limits that are achievable using combustion modification are dependent upon the fuel type (oil, coal, gas) and firing method (for coal, pulverized coal, chain-grate stoker, vibrating-grate stoker, and spreader stoker).

The cost of SO₂ control technology varies as a function of boiler size, load factor, and fuel sulfur content. Thus the uncertainty in capital and annual costs can be large. The capital costs and operating costs shown in Figures A.6.1 and A.6.2 can be in error by as much as +40 percent. The cost of retrofitting industrial boilers is highly uncertain since space limitations and other restrictions can cause significant variations.

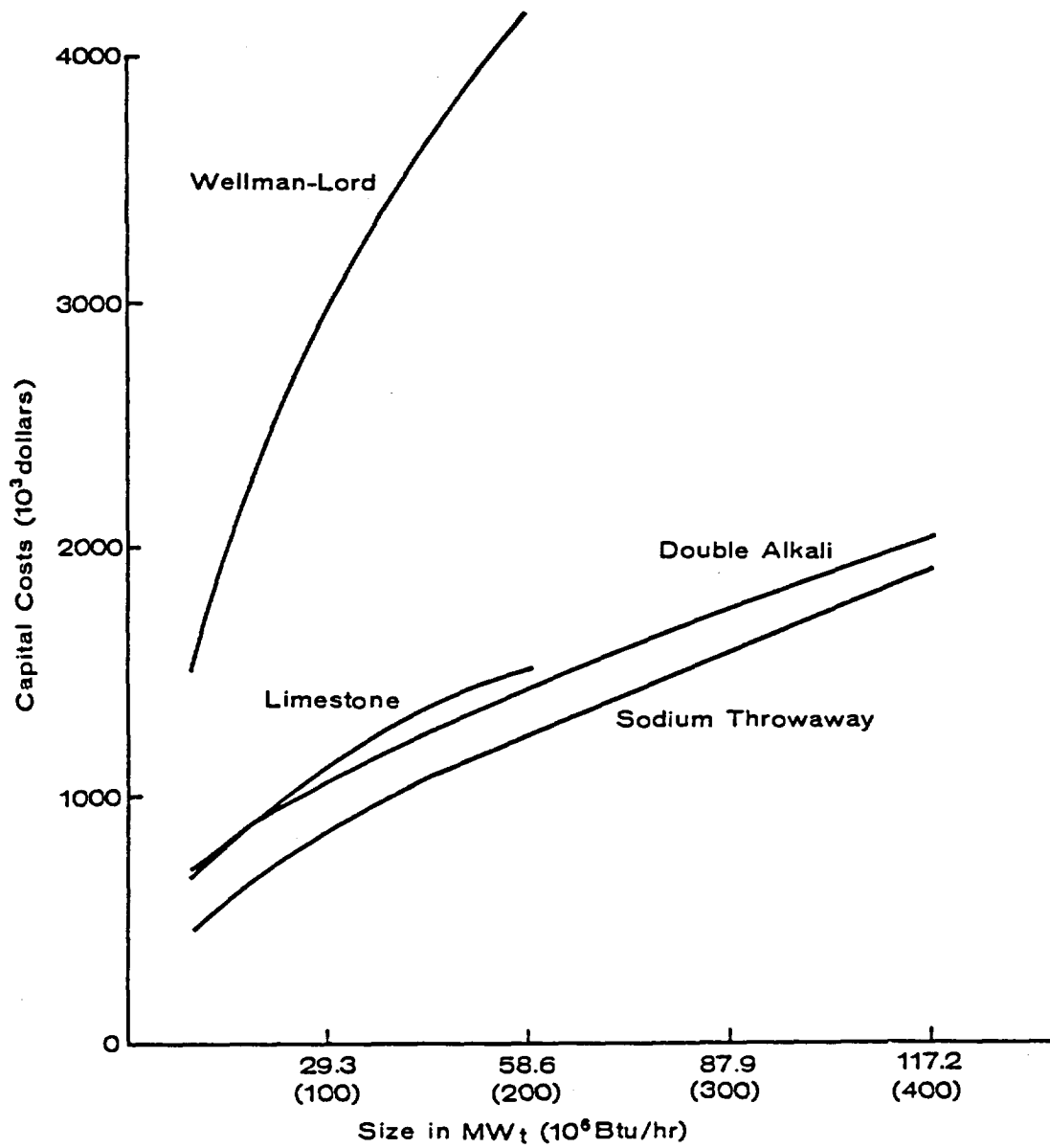


FIGURE A.6.1 FGD CAPITAL COSTS VERSUS UNIT SIZE
(3.5% S coal, 90% removal)

Source: Technical Assessment Report for Industrial Boiler Applications:
Flue Gas Desulfurization
Industrial Environmental Laboratory; USA E.P.A.
November 1979

Note: Costs are given in U.S. \$ (1978).

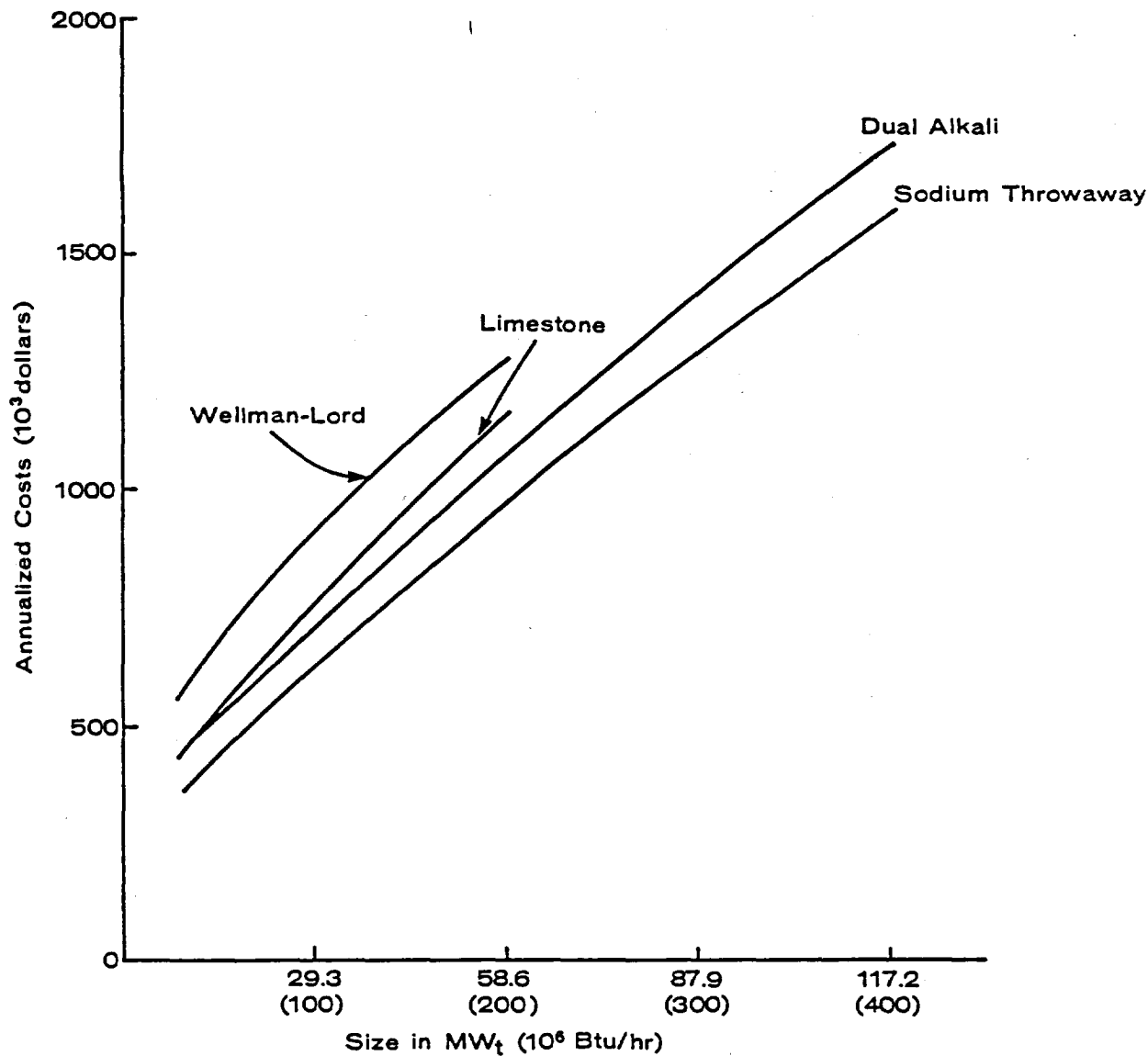


FIGURE A.6.2 FGD ANNUALIZED COSTS VERSUS UNIT SIZE
(3.5% S coal, 90% removal)

Source: Technical Assessment Report for Industrial Boiler Applications:
Flue Gas Desulfurization
Industrial Environmental Laboratory; USA E.P.A.
November 1979

Note: Costs are given in U.S. \$ (1978).

NO_x control technology for commercial and residential boilers has not progressed as rapidly as for the larger boilers, primarily because of the considerably smaller emission reduction potential for this sector. However, results of research indicate that some emission reduction is economically possible for commercial and residential boilers. Precise cost figures for these boilers are not available, but preliminary indications are that any increase in cost will be largely offset by the fuel savings and increased thermal efficiency.

A.7 RESEARCH AND DEVELOPMENT SUMMARY

The principal research and development activities currently underway are directed toward the control of sulfur dioxide and nitrogen oxides from fuel combustion processes. They can be grouped into three main areas; SO₂ control, NO_x control and combined SO_x/NO_x control.

With respect to SO₂ control, both wet and dry flue gas desulfurization technologies are being examined with a view to enhanced removal efficiencies. As well, studies are underway to assess state-of-the-art and advanced coal cleaning methodologies as techniques for meeting SO₂ emission regulations.

NO_x controls are being examined from the perspective of combustion modification techniques for a number of different fuel types.

Combined SO₂/NO_x controls are being assessed by the development and evaluation of limestone injection multistage burners (LIMB) and fluid bed combustion.

Recommendations are made in this report regarding other R & D initiatives that could be undertaken. These include further work on SO₂ and NO_x reduction for fossil-fuel-fired electrical generation processes and industrial boilers, process and control technology development projects for the control of SO₂ emissions from non-ferrous smelters and projects to improve emissions inventories.



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