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ATMOSPHERIC SCIENCES AND ANALYSIS WORK GROUP 2

UNITED STATES — CANADA

MEMORANDUM OF INTENT ON TRANSBOUNDARY AIR POLLUTION

**REPORT NO. 2F-L
NOVEMBER 1982**

WORK GROUP 2

ATMOSPHERIC SCIENCES AND ANALYSIS

FINAL REPORT
TECHNICAL BASIS

Local and Mesoscale Analysis
Subgroup Report

Subgroup Co-Chairmen

Donald Henderson, United States
Roderick Shaw, Canada

Report No. 2F - L
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Submitted to Work Group 2 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.

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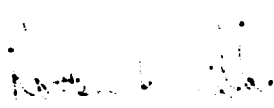
Dr. Lester Machta, Director
Air Resources Laboratory
National Oceanic and
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Dear Mr. Ferguson and Dr. Machta:

We are pleased to transmit under cover of this letter the final report of the Local and Mesoscale Analysis Sub-Group of Working Group 2, Atmospheric Sciences and Analysis, as provided for in the Phase III Work Plan. We believe that this report satisfies in a scientifically responsible manner our Phase III terms of reference.



Donald Henderson
U.S. Co-Chairman
Sub-Group on Local and
Mesoscale Analysis



Roderick Shaw
Canadian Co-Chairman
Sub-Group on Local and
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1 INTRODUCTION

Most of the emphasis in the analysis of transboundary pollution problems has been on the transport, diffusion, transformation and deposition of atmospheric pollutants over distances of the order of 1,000 kilometers (km). However, the understanding and prediction of atmospheric processes occurring in the local to mesoscale range (up to 300 km from the source) are also important for the following reasons:

(a) There are many important instances of transboundary transport of air pollution which occur over local and mesoscale distances. Examples are sulfur dioxide, oxidants, particulates and hazardous hydrocarbons in the Detroit, Michigan--Windsor, Ontario area; hydrocarbons and particulates in the Sarnia, Ontario-Port Huron, Michigan region; fluorides on Cornwall Island, Ontario, near the New York border; sulfur dioxide in the Edmundston, New Brunswick-Madawaska, Maine region; and sulfur dioxide from Poplar River, Saskatchewan, near the Montana border.

(b) There are sensitive areas such as national park lands located at or near the international boundary which could be adversely affected by pollutants undergoing local or mesoscale transport. For example, Glass and Loucks (1981) report potential effects of the proposed Atikokan power plant on the Boundary Waters Canoe Area and Voyageurs National Park in Minnesota. Stottlemeyer (1981) has limited data that indicates reduced pH in watersheds in Isle Royale National Park resulting from local and regional transport.

(c) Concentration and deposition effects from sources within 300 km augment and sometimes dominate those from the long-range transport of air pollution depending on location and meteorological conditions.

(d) With respect to emissions from a given source region, the existing LRTAP models are not always suitable for predicting concentrations and depositions within the source region itself. One reason is that these models by design do not resolve horizontal and vertical details within the mesoscale range. Another reason is that the models may not adequately represent the detailed chemistry occurring in the first few hundred kilometers

of travel. This may, in turn, have a significant effect upon the models' predictions at longer distances. Improved knowledge and predictions of mesoscale effects should, therefore, improve the predictions of the LRTAP models.

(e) When computing sulfur and nitrogen budgets using existing deposition monitoring network data, the deposition sink terms may be underestimated, since most of the existing networks in North America are of a "regional" nature, and an attempt is made to locate samplers as far away as possible from large local sources. It is important to assess the magnitude of this underestimation.

The assessment of air pollution concentrations and surface deposition, which is attributable to local sources in a given area, must include: (a) the contribution to the specific location from long-range transport; and (b) the contribution to the specific location from sources that are located within a specified distance (less than a few hundred kilometers). Being able to distinguish the local/mesoscale contribution from the long-range transport contribution is especially important to the U.S./Canada Transboundary Committee. This information is necessary to determine the most effective control measures to recommend.

This report will address two aspects of the local/mesoscale problem:

(a) A discussion of evidence of local and mesoscale transport and deposition, including transboundary flow that has been reported in the literature and in data sets. Factors affecting the importance of local effects will also be discussed.

(b) A survey of available and applicable local/mesoscale models worthy of consideration and testing.

The emphasis will be on sulfur and nitrogen compounds which lead to acid deposition. However, some discussion of ozone, particulate matter and hazardous organics will also be included.

2 EVIDENCE OF LOCAL AND MESOSCALE EFFECTS

Because different atmospheric factors may be at play, near-source effects of plumes from individual point sources will be discussed separately from the effects of urban or area sources.

2.1 Plumes from Individual Point Sources

Chimney plumes have on occasion been tracked out to several hundred kilometers, especially under "dry" conditions (Millan and Chung, 1977; Husar *et al.*, 1978; Williams *et al.*, 1980). The latter two papers discussed the dispersion of chimney plumes on the scale of interest, and have shown the importance of such factors as diurnal variations in the structure of the boundary layer and wind shear. The importance of nearby sources will depend partly upon the rate of oxidation of SO₂ to sulfates. A recent summary of data on the chemical transformation rates that occur during plume transport has been given by Newman (1980), for the case of sulfur dioxide oxidation. The following general conclusions emerge from a consideration of Newman's survey paper, and some other investigations as indicated below.

The SO₂ oxidation rate in the absence of liquid water is usually small, typically about 1% h⁻¹. Limited data indicate that, under summertime conditions, the NO_x conversion rate to nitrates is several times the SO₂-to-sulfate rate (Forrest *et al.*, 1980; Richards *et al.*, 1980). Although the exact role of heterogeneous reactions is unclear, the evidence suggests that homogeneous gas-phase processes are certainly important, and can lead to SO₂ transformation rates up to about 4% h⁻¹. The quality of the "dry" oxidation rate data so far has been such that parameterizations beyond simple first-order processes have not been justifiable, although Gillani *et al.* (1980) have recently proposed an interesting expression derived from their data from a number of power plant plumes, which seems to predict rather well SO₂ oxidation rates from solar radiation, mixing height and ambient ozone concentration data.

Chemical transformation rates under very humid conditions, of relevance during rain, are uncertain, because of the absence of direct

measurements. The only such measurements, published by Gartrell et al. in 1963, are probably flawed (Newman, 1980). However, some workers have inferred from other data that "wet" transformation rates of SO_2 can be greater than $10\% \text{ h}^{-1}$ (Dana et al., 1975; Enger and Högström, 1978; Scott, 1980).

There have been few relevant investigations of the impact of point sources on the mesoscale dry deposition field, as determined by field studies because of the difficulty in making direct dry deposition measurements. Husar et al. (1978) and Gillani et al. (1978), in their studies of the Labadie power plant plume, used plume mapping and subsequent sulfur budget estimates, and found mid-day removal rates as high as $10\% \text{ h}^{-1}$ for sulfur dioxide, with negligible values during the night when the plume is decoupled from the ground. The percentage of the power plant emissions deposited was a function of the time of release of the emissions, and could approach 50% after one day under certain conditions. These results apply to summertime conditions, and rates would be correspondingly lower in more stable, winter atmospheres (see, for example, de Wys et al., 1978). Barrie (1980) used artificial deposition surfaces to examine dry deposition patterns of sulfur and heavy metals around an isolated power plant in the summertime. He found that dry deposition processes removed only about 1% of the primary particulate sulfur and vanadium (a metal released mainly with power plant emissions) within 25 km. Wintertime total deposition measurements (i.e., dry and wet deposition) around the same plant showed that less than 0.5% of the total sulfur was removed within 25 km--results that are similar to those obtained under wintertime conditions by Summers and Hitchon (1973) in the vicinity of a sour gas plant, who found that less than 2% of the emitted sulfur was deposited within 40 km.

From the limited data discussed above, it is difficult to generalize the impact of point sources on mesoscale dry deposition. One can say, on theoretical grounds, that the relative contribution of a given source to dry deposition will depend on its emission rate, the local meteorology, the time of year and surface characteristics of the surrounding area, nature of the emissions from the source including source height and emission rate, and especially the "background" deposition from natural and anthropogenic sources. The effect of local emissions on the regional dry deposition field can be estimated by superimposing on regional background concentration levels

the modeled incremental concentrations due to the local source, because the dry deposition rate is expected to be proportional to the ambient concentration (see, for example, Granat and Söderlund, 1975). However, these estimated concentrations and depositions assume linear chemistry and, consequently, additivity of the effects of individual sources.

Somewhat more experimental information is available on wet deposition due to point sources. In examining the evidence in the literature, one must distinguish between wet removal during a precipitation event and climatological averages. For example, a high rate of removal during precipitation might result in a climatologically averaged removal of only a few percent since precipitation occurs only a small fraction of the time. Some of the available studies of precipitation scavenging of power plant, smelter and other plumes (Granat and Rodhe, 1973; Summers and Hitchon, 1973; Hutcheson and Hall, 1974; Larson et al., 1975; Granat and Söderlund, 1975; Dana et al., 1975; Wiebe and Whelpdale, 1977, Enger and Högström, 1979; Chan et al., 1981) have shown detectable effects on precipitation chemistry out to as far as 100 km. These studies suggest that smelter plume particulates are removed very efficiently during rainy days (Larson et al., 1975; Wiebe and Whelpdale, 1977; Chan et al., 1981). "Bulk" deposition (i.e., wet plus an unknown portion of dry deposition) experiments over periods of several months around the INCO smelter at Sudbury (Mueller and Kramer, 1977; Scheider et al., 1981) give similar results. The fate of the emitted sulfur is less understood, and different investigators have come to conflicting conclusions. For example, Granat and Rhode (1973), Granat and Söderlund (1979), Larson et al. (1975), Wiebe and Whelpdale (1977) and Chan et al. (1981) found that the removal of sulfur by rain from power plant and smelter plumes is a relatively inefficient process, only a few percent of the emissions being removed within about 15 to 50 km of the source even during precipitation itself. If the results were averaged over both wet and dry periods, the fraction deposited would be about an order of magnitude less. On the other hand, the results of Summers and Hitchon (1973) and Enger and Högström (1979), indicate much greater local deposition rates during a precipitation event. The results of the last two papers might be explained on the grounds that the summertime convective storms in Alberta studied by Summers and Hitchon are very efficient scrubbers for local pollution. Some of the air masses involved in Enger and

Högström's study may have contained relatively high ammonia concentrations which would have speeded up the sulfate removal. More experimental data are needed, however, to elucidate the various mechanisms involved in removal in different circumstances.

As far as the local impact of a point source on the surrounding wet deposition field is concerned, the results depend (as expected) on such factors as the strength and composition of the local emissions, and the extent to which the air mass entering the region of interest has been polluted by upwind sources. For example, Dana et al. (1975) found that the capability of a storm to wash out SO_2 from a power plant plume depends on the chemical makeup of the scavenging precipitation, since the solubility of SO_2 is very sensitive to the acidity of the absorbing raindrops (Hales, 1978). Similarly, Chan et al. (1981) found that during rainstorms the relative contribution of the INCO smelter to wet deposition in the Sudbury Basin depends on the weather system passing through the area. Wet removal of SO_2 is greater for cold frontal storms, which come from northern Canada and are significantly less acidic on arrival than the warm frontal storms associated with southerly air masses. This particular source contributed about 10 to 20% of the total wet deposition of sulfur out to 50 km or so. The smelter contribution of the total deposition of copper and nickel within the same area was roughly 40%.

For a large part of the period between June 1978 and May 1979, the Sudbury area INCO smelter was shut down due to strikes or scheduled shutdown. A detailed analysis was carried out by the Ontario Ministry of the Environment of the H^+ and SO_4^- in bulk and wet-only deposition samples collected by a network of monitoring stations in the local area (100 km). Comparisons of this data have been made with that obtained during periods when the smelter was operational. The results of this analysis did not show any significant change in the rainwater pH or acid deposition in the local area as a result of the smelter shutdown, thus providing evidence that long-range transport has a large impact on the acidic deposition in the Sudbury Basin. The comparison did show that the shutdown had a small but detectable effect on the amounts of sulfates collected within 20 km of the source. The results are of interest not only when assessing the potential impact of a large point source on the local area, but also in showing the important impact of long-range transport

of acid precursors, in that in spite of the shutdown, the precipitation in the area continued to be highly acidic. One can only surmise as to how much more acidic the precipitation would have been at distant locations downwind of the smelter, with the plant in operation, because of the insufficient density of monitoring stations in those areas.

2.2 Plumes from Urban and Area Sources

(a) Sulfates and Nitrates

The study of near-source effects from urban and area sources is complicated by many factors. Among these are the multitude of many sources emitting different pollutants at different heights which interact chemically to form secondary pollutants, the large spatial extent and dispersed nature of the urban/area source, and the effect of an urban area upon the local meteorology, such as increased vertical dispersion due to increased surface roughness and possible modification of precipitation patterns due to the injection of condensation nuclei.

As is the case with plumes from single point sources, there appears to be conflicting evidence in the literature about the magnitude of near-source effects from urban/area sources. Some authors have found that little of the sulfur or nitrogen emitted by an area source is deposited within the first 100 km or so of travel. For example, Rodhe (1970) examined total sulfur content in snow samples in February 1969, around Uppsala, Sweden, and came to the conclusion that only about 3% of the sulfur emitted in the town during a 12-day period was deposited within 3 km, and less than 5% within 15 km. However, because of the low background values, the deposition within 5 km of the town was three or four times the regional levels. Rodhe *et al.* (1972), examined daily concentrations of soot and sulfate aerosols in the vicinity of 10 towns in southern Sweden during a four-month period and found that, during that period, only 5% of the emitted SO_2 had been oxidized to aerosol sulfate within the town (i.e., during a few tens of minutes of travel). They estimated that locally produced sulfate accounted for less than 50% of the total sulfate measured in the towns.

Davies (1976), found that the precipitation scavenging of SO_2 in a 12 km^2 area around the industrial city of Sheffield, U.K., was quite small in that only 0.3% of the emitted SO_2 was removed during precipitation. However, sulfate in the precipitation was not measured and may have accounted for a much greater fraction of emitted sulfur being deposited in the industrial area.

Högström (1979), examined in a theoretical manner the dry deposition of sulfur in the first few kilometers downwind of an urban source, before complete mixing in the vertical had been established. He came to the conclusion that, because of the much more rapid rate of vertical dispersion over the relatively rough surface of a large city compared to that in a rural area, the proportion of sulfur dioxide deposited locally should be larger for a small city than for a large one. From measurements of SO_2 at Vert-le-Petit, 37 km south of Paris, he concludes that only a small fraction (5 to 10%) of SO_2 emitted in Paris was deposited during the first few kilometers of travel. It should be stressed, however, that Högström's study dealt with dry, and not wet, deposition.

Benarie (1976) used measured concentrations of SO_2 and sulfate, also at Vert-le-Petit, to estimate that 50% of the sulfur emitted in Paris is deposited within a 37 km circle surrounding the city. The difference between this estimate and that of Högström (1979) has sparked an ongoing debate in the literature. Benarie's analysis assumed vertical and cross-wind uniformity of SO_2 and sulfate concentrations in the plume, and that the size of and concentrations in the Paris plume as it passed over Vert-le-Petit were representative of all wind directions. In addition, the budget calculations are highly dependent upon the value assumed for the deposition velocity. Nevertheless, the approach has possibilities and should be extended with monitoring stations in all directions from a major urban or industrial source.

Other studies also seem to indicate that local emissions may be important. Many of these studies have taken place at the same locations where some of the above-mentioned studies have indicated a small local effect. For example, in a 400 km^2 area around Uppsala, Sweden, Andersson (1969) examined the deposition by rain of S, H^+ , Ca, Na, K, and Cl from July to October 1962,

and found that the maximum concentration in the collected precipitation increased several-fold over regional background values and was located downwind of the city. No estimate was made, however, of the fraction of locally emitted material that was deposited locally and, therefore, this study does not contradict that of Rodhe (1970) referred to above. However, in a later paper, Högström (1974) measured the wet deposition of sulfate at 100 sampling points within 60 km of Uppsala and estimated that two-thirds of the sulfur emitted during precipitation was deposited within 50 to 100 km of the source. He also concluded that 98% of the emitted sulfur was rapidly oxidized and deposited as sulfate during the precipitation events. Of course, the long-term average would be much lower. It should be pointed out that Uppsala (population approximately 100,000) is a rather unusual urban source, not typical of a North American city, in that approximately 65% of the SO₂ emitted in that city comes from a single district heating plant. The height of the stack, however, was only 40 m.

In the vicinity of Sundsvall, Sweden, approximately 30 lakes have been judged to be sensitive to further deposition of atmospheric acid. A survey of the sulfur content of snow samples in the region indicated that emissions from smelters within a few kilometers contribute greatly to the deposition. The observed pattern of deposition coincides almost exactly with the lakes that have been affected most by acidification (Sundsvalls Kommun, 1980).

Esposito et al. (1980) propose that in the northeastern United States, primary emissions of sulfate from the combustion of residual oil in power plants and, to a greater extent, in commercial and residential boilers, and the accompanying emissions of vanadium and nickel catalysts are a major cause of acid precipitation. These authors also contend that primary sulfate from oil-burning-power plants and oxides of nitrogen from local motor vehicle exhausts are an important factor in the acidification of precipitation in Florida and in California. However, their thesis is based upon emission estimates and a box model, the chemical processes in which are not described in their paper, and needs to be further supported by more detailed evidence and by meteorological studies such as back-trajectory analyses. Shannon (1981) used his statistical trajectory model to estimate the relative impor-

tance of primary sulfate along the Atlantic seaboard of the United States. During winter the model estimates that more than 50% of the ambient ground-level sulfate concentrations is due to primary sulfate. This is because there are large emissions of primary sulfate from heating units that have low stacks, and also because the production of secondary sulfate is slower in the winter.

Hales and Dana (1979), sampled summer precipitation in the vicinity of St. Louis during the METROMEX Project. When normalized to the precipitation amount, the sulfate and nitrate concentrations in samples taken during individual storms showed well-defined maxima downwind of the city. The deposition of sulfate and nitrate during convective storms, as measured by the network, were comparable in magnitude to emission rates of SO_x and NO_x from the St. Louis urban source. The data seemed to indicate that reactive scavenging of locally emitted SO_2 , rather than scavenging of regional sulfate, was the important mechanism.

In Nova Scotia, Shaw (1982) sampled aerosol sulfate and precipitation during a one-year period at a site 25 km from Halifax, a city of 300,000. Through an analysis of back-trajectories and wind patterns, he concluded that 40 to 50% of the deposition of sulfur and hydrogen ion at the site during precipitation were due to emissions from Halifax; the deposition during precipitation was consistent with a removal coefficient for sulfur of $2 \times 10^{-4} \text{ s}^{-1}$ or about $70\% \text{ h}^{-1}$. On an annual basis, these results indicate that about 10% of the sulfur emitted in Halifax is deposited by precipitation within 25 km of the city. Watt et al. (1979), found a significant increase of hydrogen ion and sulfate concentrations in lake water in the vicinity of Halifax as they sampled closer to the city, implying that sources in Halifax were an important factor contributing to the concentrations.

Wiltshire (1979), carried out a sulfur budget for Nova Scotia by analyzing actual data for total deposition and concentration and comparing them with deposition from Nova Scotian sources as estimated by a simple model. He concluded that, on the average, 25% of the total (wet plus dry) deposition of sulfur in Nova Scotia is due to emissions from Nova Scotian sources. Furthermore, 30% of Nova Scotian emissions are deposited within that

province. More recent and spatially detailed modeling work by Wiltshire (1981), shows well-defined maxima of deposition (as estimated by his model) in the vicinity of the strong urban and industrial sources of Atlantic Canada, such as Halifax, Saint John and Port Hawkesbury.

The results that have been summarized above seem to be contradicting. Part of the confusion arises from the fact that the experimental data have been interpreted in different ways. The influence of local sources has been interpreted in terms of (1) increase in local concentrations and deposition over regionally averaged values; and (2) fraction of emitted material deposited during precipitation and of fraction of emissions deposited in the long term. It is, therefore, important to be clear about the way in which local and mesoscale influences are expressed. Most of the papers referred to above seem to say that local sources can increase concentrations in air and precipitation within 10 to 20 km appreciably above regionally averaged values. Most also seem to agree that, on the long term, local sources account for a few, or at most 10 to 25%, of the deposition within a few tens of kilometers of the source. On the other hand, the most disagreement among researchers is with respect to the fraction of the local emissions that is deposited during precipitation; this fraction can vary from a few to virtually 100%. This range of estimates may result from data interpretation, or it may be caused by different factors, both physical and chemical, which are at play at different times and locations. What is needed are more field studies of local and mesoscale effects and perhaps a re-analysis of past studies if future studies shed new light on the matter.

Several workers have discussed the factors affecting the residence time or fallout scales of atmospheric pollutants, especially sulfur. Bolin et al. (1974) have discussed in a theoretical way some of the meteorological factors governing the residence time of pollutants in the atmosphere. A short residence time would imply that pollutants are removed from the atmosphere close to the source. The residence time increased markedly with decreasing deposition velocity and only slightly with increasing height of emission into the atmosphere. The residence time increased with increasing surface roughness. Wind speed was not of great importance. They concluded that dry deposition was likely to be less important than incloud scavenging. Indeed,

Högström (1979) found that dry deposition would deposit only about 5 to 10% of the sulfur emitted by a city within a 70 km travel distance, for deposition velocities in the reasonable range of 0.8 to 1.6 cm s⁻¹. In agreement also with Bolin et al. (1974), he found that increasing city size (increasing roughness) led to a decrease in local deposition (or an increase in residence time) because the increased mechanical turbulence would lead to greater vertical dilution, decreased concentrations near the surface and, therefore, slower dry deposition.

Removal by precipitation has the same effect as a large increase in deposition velocity or removal rate (McMahon and Denison, 1979). The probability of emitted sulfur being deposited by wet processes within a few hours' travel of the source will depend upon the probability of the sulfur being irreversibly absorbed into cloud and/or rainwater and the probability of precipitation occurring during the initial travel. Rodhe and Grandell (1972) estimate, by taking into account the mean length of wet and dry periods, that the lower limit of the average turn-over time of sulfur in Europe is of the order of 50 hours.

Högström (1978) used a simple model and observations of sulfur in air and precipitation to estimate the wet fallout parameter k in Sweden, i.e., the fraction of pollutant removed per unit time by precipitation. Interestingly, k varied geographically. Over much of southern Sweden, it had a value of 1 to 2 x 10⁻⁴ s⁻¹ (corresponding to 2/3 fallout within 50 to 100 km from the source) but, along a strip 100 km wide along the west coast, k had a value of approximately 2 x 10⁻⁵ s⁻¹ (corresponding to 2/3 fallout within 500 to 1,000 km). His estimates were in good agreement with the observational studies of Högström (1974), Enger (1977) and Granat and Söderlund (1975). In his opinion, inaccuracies in air quality and precipitation chemistry data, emission estimates, or meteorological data could not account for the geographical variation in the estimates of k. Högström (1974) alludes to the possibility that the chemical composition of the air is the cause of the geographical variation; the chemical makeup of cloud and rainwater will have a pronounced influence on the rate of oxidation of sulfur dioxide.

It would appear from the above discussion, therefore, that local effects from area sources (and possibly point sources) may vary a great deal geographically and temporally in that they may be quite episodic. The study of Shaw (1982), indicates that, during a one-year period, about 10% of sulfur emissions from Halifax were deposited within a 25 km radius of the city. During individual storms, the deposition within 25 km was of the same order as the emissions. Similarly, Hales and Dana (1979), observed that, during summer convective storms, the deposition of SO_4^- and NO_3 to the observational network in the vicinity of St. Louis was comparable in magnitude to the emissions of SO_x and NO_x from that urban source. It appeared that the pollutants in the urban plume were very efficiently removed. Hales and Dana suggest that this may have been due to aqueous phase oxidation of SO_2 to SO_4^- by oxidants such as ozone and hydrogen peroxide, with assistance from catalytic metals that may be found in the urban plume. Penkett *et al.* (1979), concluded that O_3 and H_2O_2 were potentially the most important aqueous phase oxidants for SO_2 and that, in the absence of catalysts, oxidation by H_2O_2 would dominate over that by O_3 for droplet pH values of less than 5. Conversion of SO_2 to sulfate in cloud water would, in contrast to the dissolution of SO_2 alone, amount to an irreversible removal of SO_2 . Similarly, rapid conversion of NO and NO_2 to NO_3^- may be due at least partly to photochemical oxidants in the urban plume.

It is possible that alkaline pollutants may offset the acidification taking place in cloud and raindrops. Kemmerer and Jackson (1973), observed alkaline rain in the immediate vicinity of Syracuse, New York, and concluded that calcium oxide and other neutralizing particulates from industrial and incinerator emissions were causing the pH to be higher near the city than at the rural sampling sites.

As far as oxidation of sulfur dioxide in urban plumes is concerned (in the absence of precipitation), Alkezweeny (1980) has given a good summary of most of the available data. These data suggest that oxidation rates in urban plumes are somewhat higher than those in chimney plumes, possibly by a factor of two or more (values greater than $10\% \text{ h}^{-1}$ have been reported by several workers), and that heterogeneous mechanisms may play a greater role in urban than in chimney plumes. Little data is available on nitrogen oxides

transformation rates in urban plumes. Spicer (1979) has reported summertime values of over $20\% \text{ h}^{-1}$, most likely due to homogeneous reactions.

(b) Oxidants

The long range transport of oxidants and their precursors, which causes high oxidant episodes, is usually associated with the presence of high pressure systems. This report will not consider long-range oxidant transport that is discussed in the Monitoring and Interpretation Subgroup report, but instead will restrict comments to the impact of urban plumes on oxidant levels up to distances of the order of 300 km. Urban plume studies relative to oxidants have been conducted in a number of areas, including New York, Chicago, Boston, Philadelphia, Tulsa, St. Louis, the southern California urban area, and Toronto.

Ludwig et al. (1977) report that maximum oxidant levels may occur as far as 150 km from the city centre. Spicer et al. (1979), report urban plumes of 80 to 128 km in width and 180 to 280 km in length. Spicer also reported the approximate maximum contribution of various sources to the ozone concentration. Ozone generated within urban plumes contributed 150 to 200 ppb on an hourly average. Eaton et al. (1979) concluded that the net ozone formation in the Tulsa plume resulted in concentrations of 17 to 116 ppb; maximum ozone concentrations were observed to occur from 30 to 50 km downwind of the urban area. Wolff et al. (1977) reported that the maximum increase in ozone concentration associated with its generation in an urban plume was 78 ppb, observed 60 km downwind of the northeastern New Jersey-New York area. Predictions by the photochemical model of Bazzell and Peters (1981) show a net increase in ozone of approximately 70 ppb in the urban plume. Chung (1977) estimates from observations that Toronto produces up to 60 ppb in its downwind urban plume.

Ozone concentrations generated in urban plumes tend to be greater at larger distances when the plumes are transported over large bodies of water because of less diffusion, particularly in the vertical direction under relatively stable stratification. Spicer et al. (1979), Cole et al. (1977), Blumenthal (1976), Chung (1977), Lusi et al. (1976) and Westberg et al.

(1976), have all reported on oxidant levels with respect to transport over water and/or sea or lake breezes. The southern Great Lakes area may be especially important because of the appreciable NO_x concentration (Ludwig et al., 1977).

There remains a need for a better understanding of oxidants, especially with respect to their formation and relationship to precursor chemicals in the urban and rural areas. Several authors (Martinez et al., 1979; Jefferies et al., 1976; Dimitriades, 1977; Singh et al., 1978) caution that control strategies established for urban areas may have little benefit or perhaps even detrimental effects on rural oxidant levels. Consequently, control strategies that might be implemented in an urban area of one country might increase ozone levels in rural areas of the other country.

Indications of transboundary movement of ozone into Ontario have been documented by Chung (1977) and Shenfeld et al. (1978). Mukammal (1964) reported on the relationship between "weather fleck" on tobacco and meteorological conditions associated with the buildup of ozone in an area immediately north of Lake Erie.

Since 1977, annual reports to the U.S.-Canada International Joint Commission (IJC) by the International Michigan-Ontario Air Pollution Board have commented on the long-range transport of ozone and precursor pollutants into the boundary area along the Detroit and St. Clair Rivers. The high ozone levels were noted to be associated with warm humid air moving into the area from the south and southwest directions. There is evidence that local emissions add to the levels of ozone downwind of the sources in the boundary area.

(c) Suspended Particulate Matter

Much of Detroit and Windsor experience unacceptably high levels of total suspended particulates. The Joint Air Pollution Study of St. Clair-Detroit River areas for the International Joint Commission (January, 1971), reported that based on a 1968 study "The dispersion model estimates show that the combined contribution of U.S. area and point sources to the annual average

concentration of particulates in the Windsor area were very significant. U.S. sources contribute the equivalent of at least the entire annual average particulate concentration loadings allowed under Ontario standards ($60 \mu\text{g}/\text{m}^3$) for a large portion of the Windsor area. For some sections of the area, particulate pollution from the U.S. exceeds $140 \mu\text{g}/\text{m}^3$."

The International Michigan-Ontario Air Board report for 1980 revealed that for much of the Detroit-Windsor area, the IJC objectives for total suspended particulates were still exceeded even though there has been significant decreases in ambient levels of average TSP values annually from 1971 to 1974 and again in 1980. From 1975 to 1979, there was no significant change in annual averages of TSP. There was a very appreciable decrease in particulate emissions in Wayne County, Michigan, during 1980, much of which was attributable to a slower economy. It is in this county that 90% of the particulate emissions in the Detroit-Windsor boundary area emanate.

The Ontario Ministry of the Environment annual reports on Ambient Air Quality in Windsor, Ontario, showed correlation between elevated levels of total suspended particulates and winds from the direction of heavily industrialized areas of Wayne County, Michigan. Munn (1974) reported the transport of suspended particulate matter from the Detroit-Windsor area northward across Lake St. Clair into the Sarnia, Ontario-Port Huron area, a distance of about 100 km.

(d) Hazardous Organics

The Joint Air Pollution Study of St. Clair-Detroit River areas for the International Joint Commission (January, 1971), reported benzo(a)pyrene [B(a)P] levels in total suspended particulate samples collected in the Port Huron, Windsor and Detroit areas during 1967 and 1968. These analyses were conducted because of the carcinogenicity of polycyclic hydrocarbons. The results showed elevated levels of B(a)P in parts of Windsor and Detroit. The Windsor levels are similar to those reported in cities that have steel mills. Since there are no steel mills in Windsor the suspected origin of the elevated B(a)P are the steel mills in Wayne County, Michigan.

3 SURVEY OF LOCAL AND MESOSCALE MODELS

The Phase II report of Work Group 2 of the U.S.-Canada Transboundary Team includes a detailed investigation of long-range transport models. The investigation includes a survey of models thought to be appropriate for estimating transport, diffusion, transformation, and deposition of air pollutants over long distances. In addition, these models were used to generate transfer matrices for the purpose of determining source-receptor relationships in the United States and Canada.

As emphasized in the introduction of this report, estimates made with these long-range transport models have not demonstrated good agreement with measurements in or near the vicinity of the sources. A comparison of the various model estimates also indicates disagreements amongst the model predictions. Possible reasons for these variations are: (1) inadequate spatial resolution to accommodate near-source region contributions to air pollution concentrations; (2) differences in interpretation of the emissions inventory; (3) differences in vertical resolution and treatment of vertical diffusion; (4) inability of the models to simulate detailed chemistry close to the sources; and (5) variations in treating deposition processes.

Therefore, an attempt is made to include in this report a survey of local and mesoscale models that may be applicable to distances of the order of 300 km. The following discussion will be divided into two parts: a) local (less than 50 km); and b) mesoscale (50 to 300 km).

3.1 Local Models

Numerous publications are available in the literature that survey and review various types of models that may be appropriate out to distances less than 50 km. The majority of these models, especially the simpler Gaussian models, do not include provisions for simulating chemical transformation, dry deposition, wet deposition, and non-steady state meteorological conditions. Because many references are available in the literature concerning these models, only a brief discussion of recent publications is provided in this report. This should not be interpreted to

mean that the local transboundary transport of air pollution is insignificant, but merely to indicate that the Work Group thought that sufficient information is available in the literature to allow a user to select an appropriate local model to fit a particular application.

An excellent model review paper, Turner (1979), provides a historical perspective of atmospheric dispersion models, many references to particular models, and includes an appendix that lists those models contained in the "User's Network for Applied Modeling of Air Pollution" (UNAMAP). Turner's review paper is commented on by peers in a later paper (Egan et al., 1979). A more recent model survey by Liu et al. (1982) includes an evaluation of existing atmospheric dispersion models appropriate for estimating air pollution concentrations from elevated point sources. Thirty existing plume models were evaluated and analyzed. These consisted of 19 kinematic models, 9 first-order-closure models, and 2 second-order-closure models. The model formulation and the technical attributes of each of the 30 plume models are compared and examined for differences and similarities. A sensitivity analysis was carried out on the Gaussian plume models to explore systematically what effect varying the different algorithms would have on the overall model predictions. This sensitivity analysis will assist a user in deciding which model is most appropriate for a particular application. References to the models are given at the end of each section on the specific model types.

A steering committee, made up of United States scientists, which was organized through a cooperative agreement between the U.S. Environmental Protection Agency (EPA) and the American Meteorological Society, reviewed EPA's local dispersion models and modeling procedures. The steering committee report (American Meteorological Society, 1981) may be considered representative of current opinions on application of local models. The report recommends: (1) that EPA strongly endorse the use of atmospheric dispersion models for assistance in making decisions related to the management of air quality; (2) that EPA continue to enhance research and development activities to increase our understanding of the following topics (not listed in order of priority):

- (a) long-range transport and transformation of pollutants thought to be involved in acid precipitation,
- (b) dispersion in regions of mountainous terrain,
- (c) modeling of chemically reactive pollutants of concern to the formation of ozone and other important atmospheric materials,
- (d) modeling of the atmospheric transport, deposition, and ultimate fate of toxic materials in the ecosystem, and
- (e) identification of representative meteorological information for consideration of long-term off-site climatology and local short-term meteorology;

and (3) that EPA adopt a specific set of measures to evaluate the performance of new and existing models statistically.

3.2 Mesoscale Models

A survey of dispersion models considered appropriate for the intermediate range, 50-300 km, is provided in the following sections of this report. Systems Applications, Incorporated (SAI) surveyed 42 regional-scale models for the National Park Service (Stewart *et al.*, 1982) that may be appropriate for use in this intermediate range. In addition to the more common basic attributes of long-range transport models, i.e., transport and diffusion, physical and chemical transformation, and wet and dry deposition, SAI considered other features important for intermediate-range regional models in their model survey. These other significant features include model capability of: (1) resolving vertical pollutant distributions, (2) accommodating urban emissions, and (3) simulating detailed chemistry. A discussion of these attributes and the 42 models surveyed follows with tables included for easy comparisons of the various models and their particular attributes. Additional attributes of these models may be important to consider in assessing their suitability for particular applications. These include input data requirements (meteorology, terrain, emission source details), computer requirements, resolution of predicted concentration and deposition fields. These attributes were not considered here because the details of intended applications are not known.

(a) Vertical Resolution of Pollutant Distributions

The vertical distribution of pollutants is generally more important for model applications at intermediate-range distances; therefore, the majority of the long-range transport models do not include attempts to resolve them. However, some regional models (both long-term and episodic), which include vertical resolution, do exist and these are more suitable for intermediate-range modeling provided that data input resolution is adequate for these scales. From the survey of the 42 regional models each was categorized according to its vertical resolution treatment. This information is presented in Table 1. No attempt has been made to rank models within each category.

(b) Accommodation of Urban Emissions

A recent emissions inventory compiled for the EPRI/SURE region (Klemm and Brennan, 1981) indicates that minor point sources and area sources (residential, commercial, industrial, and transportation) contribute 17% of the total sulfur emissions, 62% of the total nitrogen emissions, and 56% of the total particulate emissions. A large majority of these emissions occur in urban areas. In order to treat urban emissions adequately, the model must provide for area source emissions and surface-based emissions.

None of the 42 models surveyed specifically focused on urban emissions, and very few mentioned these emissions at all. This is not surprising because the primary concern was sulfur emissions over large spatial scales. Over intermediate distances, urban influences play a significant role, depending on the particular pollutant being considered. Some of the models have the capability to treat urban emissions with little or no modification. These are the Eulerian grid models, and more specifically, the grid models with some vertical resolution.

Lagrangian models may easily incorporate urban emissions if they are receptor-oriented. In these models, emissions are gridded and input into a trajectory segment, or air parcel, as it flows toward a receptor. However,

Table 1 SURVEY OF REGIONAL AIR QUALITY MODELS SUITABLE FOR INTERMEDIATE RANGE APPLICATION

No Vertical Resolution	Moderate Vertical Resolution	Sufficient Vertical Resolution
Uniform distribution	Ground level estimates distinct from mixed layer; or Gaussian distribution in vertical (reflections or no reflections); or Gaussian distributions modified by dry deposition; or Other analytic expression for vertical distribution	Vertical diffusion equation integrated; or Variable K_z profiles; or Multiple-layer models
<p>AES/LRT^{2,4,7} (Voldner et al., 1980) CAPITA/MCARLO^{3,12} (Patteraon et al., 1980) CCIW/LDAPM^{2,8} (McMahon, 1976) DMI/LRPDM^{1,10} (Prahm and Christenaen, 1977) ERT/ATM^{1,4,7} (Rao et al., 1976) IAP/LRT^{1,4,7} (Szepesi, 1978) NIAR/RTM^{1,4,7} (Eliassen and Saltbones, 1975) OME/LRT^{2,5,6} (Venketram et al., 1980) SCAR/LRTM^{1,4} (Nordlund, 1975)</p>	<p>ARL/ATAD^{3,4,6} (Heffter, 1980) CERL/LTSDM^{2,6,8} (Fisher, 1978) ERT/MESOPLUME^{1,4,6} (Berkley and Bass, 1979a) ERT/MESOPUFF^{1,4,6} (Berkley and Bass, 1979b) MEP/LRT^{2,5,6} (Weisman, 1980) PNL/LRT^{3,4,6} (Wendell et al., 1976) PNL/RPTM^{2,4,6} (Powell et al., 1979) PNL/STRAM^{1,4,6} (Halea et al., 1977) RI/MTDIS^{1,4,6} (Wang et al., 1980)</p>	<p>ANL/STRAP^{2,5,6} (Sheih, 1977) ANL/ASTRAP^{2,5,6} (Shannon, 1979) ARL/MTDF^{1,4,6} (Draxler, 1979) BNL/AIRSOX^{1,4,6} (Meyera et al., 1979) EPA/RAQSM^{1,11,13} (Lamb, 1980) DM/RADM^{1,4,6} (Runchal, 1980) DU-NCAR-BNL/LAMPS^{1,11,14} (Kreitzberg and Leach, 1978) ERT/SURAD^{1,11} (Lavery et al., 1978) ERT/SULFASD^{1,11} (Egan et al., 1976)</p>

Table 1 (Concluded)

No Vertical Resolution	Moderate Vertical Resolution	Sufficient Vertical Resolution
SRI/ENAMAP-1 ^{2,4,6} (Bhumralkar et al., 1980)	SAI/RTM ^{1,10} (Durrant et al., 1979)	ERT/MESOGRID ^{1,11} (Morris et al., 1979)
SRI/EURMAP-1 ^{2,4,6} (Johnson et al., 1978)	SRI/EURMAP-2 ^{1,4,6} (Mancua et al., 1979)	LLL/ADPIC ^{1,12} (Lang, 1978)
TRI/REGMOD ^{1,10} (Niemann et al., 1980)	SRL/SPM ^{1,4,6} (Kern, 1975)	SAI/ROM ^{1,11} (Wojcik et al., 1978)
TRI/RCDM ^{1,9} (Niemann et al., 1980)	SRL/PFPL ^{1,4,6} (Garrett and Murphy, 1981)	UoFS/RDDM ^{2,5,6} (Bolin and Pearson, 1975)
UMACID ^{3,4,7} (Samson, 1980)		UWATM-SOX ^{3,11} (Ragland and Wilkening, 1980)
		UoFK/RTM ^{1,11} (Carmichael and Peters, 1979)

¹Short term²Long term³Short and long term⁴Lagrangian⁵Statistical Lagrangian⁶Source oriented⁷Receptor oriented⁸Statistical⁹Eulerian analytic¹⁰Eulerian 2-D¹¹Eulerian 3-D¹²Quasi-Lagrangian, quasi-Eulerian¹³Not operational¹⁴No pollutants treated presently

the lack of vertical resolution in the models makes them less applicable to intermediate-range assessments.

Source-oriented Lagrangian models may have adequate or sufficient vertical resolution, but the puff or plume segment dimensions and dispersion rates normally pertain to point sources. Many of these models would require considerable modifications to make them useful for intermediate-range urban plume applications.

An examination of the characteristics of the 42 models, in light of the above discussion, allows a classification of the models according to whether they are (a) ideally suited to treating urban emission, (b) suitable, but perhaps with insufficient resolution for intermediate scales, and (c) not suitable without considerable modifications or numerous emissions simulations. Table 2 shows the models categorized in this manner. Note that the classification neglected the particular pollutants treated (SO_x , NO_x , HC, etc.).

(c) Simulation of Detailed Chemistry

The majority of regional and intermediate-scale air quality models focus on sulfur chemistry or simulate tracer species, or radioactive species only. However, a number of mesoscale and regional-scale photochemical models have been developed, and recent efforts to model the chemical composition of rain have resulted in the addition of simplified NO_x chemistry to those models primarily designed for sulfur treatment. A recent comparison of two photochemical oxidant models, and their sensitivity to input assumptions, may be found in Hov and Derwent (1981). This paper also contains a useful list of references.

The survey of 42 regional-scale models (SAI, 1982) has provided information from which a list has been prepared showing the level of sophistication of the chemical mechanism treated in each model. This is presented in Table 3.

A similar review of operational models that describe the transport, diffusion, and chemical transformation of air pollutants on a horizontal

Table 2 URBAN EMISSION TREATMENT

Models Ideally Suitable for Treatment of Urban Emissions ^a	Models Moderately Suitable for Treatment of Urban Emissions ^a	Models Requiring Modification for Treatment of Urban Emissions ^a
Area source capability and vertical resolution for adequate treatment of surface level emissions	Area source capability but limited or no vertical resolution	No area source capability.
EAP/RAQSM DM/RADM DU-NCAR-BNL/LAMPS ^b ERT/SURAD ERT/SULFA3D ERT/MESOGRID LLL/ADPIC SAI/ROM UWATH-SOX UoFK/RTM	AES/LRT CAPITA/MCARLO CCIW/LDAPM DMI/LRPDM ERT/ATM IAP/LRT NIAR/RTM SAI/RTM SCAR/LRTM SRI/EURMAP-1 ^d SRI/EURMAP-2 ^d SRI/ENAMAP-1 TRI/REGMOD UMACID	ANL/STRAP ^c ANL/ASTRAP ^c ARL/ATAD ^c ARL/MTDF BNL/AIRSOX CERL/LTSDM ^c ERT/MESOPLUME ERT/MESOPUFF MEP/LRT ^c OME/LRT ^c PNL/LRT ^c PNL/RPTM ^c PNL/STRAM RI/MTDDIS SRL/SPM SRL/PFPL TRI/RCDM ^c UoFS/RDDM ^c

^a See Table 1 for model reference and model type.
^b No pollutants treated presently.
^c Long-term assessments may render these models marginally suitable.
^d Source area initialized same as grid area.

Table 3 TREATMENT OF CHEMISTRY IN MODELS SURVEYED*

Model	Detail of Chemistry
AES/LRT	Linear $\text{SO}_2 \rightarrow \text{SO}_4$, seasonally varying rate; also, $\text{NO} \rightarrow \text{NO}_2$ instantaneous
ANL/STRAP	Linear $\text{SO}_2 \rightarrow \text{SO}_4$, constant rate
ANL/ASTRAP	Linear $\text{SO}_2 \rightarrow \text{SO}_4$, diurnally and seasonally varying rate
ARL/ATAD	Linear decay of pollutant, constant half-life
ARL/MTDF	No chemistry reported
BNL/AIRSOX	Linear $\text{SO}_2 \rightarrow \text{SO}_4$, constant rate
CAPITA/MCARLO	Linear $\text{SO}_2 \rightarrow \text{SO}_4$, constant rate; also, $\text{NO} \rightarrow \text{NO}_2 \rightarrow \text{HNO}_3$ and PAN chemistry
CCIW/LDAPTM	Implicit $\text{SO}_2 \rightarrow \text{SO}_4$, via half-lives of SO_2 and SO_4
CERL/LTSDM	Linear $\text{SO}_2 \rightarrow \text{SO}_4$, rate depends on downwind distance
DM/RADM	Radioactive decay, and linear SO_2 to SO_4 transformation
DMI/LRPDM	SO_2 decay only
DU-NCAR-BNL/LAMPS	No pollutants treated
EPA/RAQSM	Multispecies photochemical mechanism
ERT/ATM	Linear $\text{SO}_2 \rightarrow \text{SO}_4$, constant rate
ERT/SURAD	SO_2 oxidation based on photochemical mechanism
ERT/SULFA3D	Linear $\text{SO}_2 \rightarrow \text{SO}_4$, constant rate
ERT/MESOPLUME	Linear $\text{SO}_2 \rightarrow \text{SO}_4$, constant rate
ERT/MESOPUFF	Linear $\text{SO}_2 \rightarrow \text{SO}_4$, constant rate
ERT/MESOGRID	Linear $\text{SO}_2 \rightarrow \text{SO}_4$, constant rate
IAP/LRT	Linear $\text{SO}_2 \rightarrow \text{SO}_4$, humidity-dependent rate
LLL/ADPIC	Capability for chemistry exists but none published
MEP/LRT	Linear $\text{SO}_2 \rightarrow \text{SO}_4$, seasonally varying rate; linear $\text{NO}_2 \rightarrow \text{NO}_3$ ($\text{NO} \rightarrow \text{NO}_2$ instantaneous)
NIAR/RTM	Linear $\text{SO}_2 \rightarrow \text{SO}_4$, constant rate
OME/LRT	Linear $\text{SO}_2 \rightarrow \text{SO}_4$, constant rate
PNL/LRT	Linear $\text{SO}_2 \rightarrow \text{SO}_4$, constant rate
PNL/RPTM	Linear $\text{SO}_2 \rightarrow \text{SO}_4$, constant rate
PNL/STRAM	Linear $\text{SO}_2 \rightarrow \text{SO}_4$, function of humidity, time of day, hydrocarbon levels (4 categories)
RI/MTDDIS	Linear decay of pollutant
SAI/RTM	Linear $\text{SO}_2 \rightarrow \text{SO}_4$, dependent on latitude and time of day
SAI/ROM	Multispecies photochemical mechanism
SCAR/LRTM	Constant decay rate of SO_2
SRI/ENAMAP-1	Linear $\text{SO}_2 \rightarrow \text{SO}_4$, constant rate
SRI/EURMAP-1	Linear $\text{SO}_2 \rightarrow \text{SO}_4$, constant rate

Table 3 (Concluded)

Model	Detail of Chemistry
SRI/EURMAP-2	Linear $\text{SO}_2 \rightarrow \text{SO}_4$, constant rate
SRL/SPM	Radioactive decay rate
SRL/PFPL	Radioactive decay rate
TRI/REGMOD	Linear $\text{SO}_2 \rightarrow \text{SO}_4$
TRI/RCDM	Linear $\text{SO}_2 \rightarrow \text{SO}_4$, constant rate
UMACID	Linear $\text{SO}_2 \rightarrow \text{SO}_4$, diurnally varying rate
UofK/RTM	Homogeneous $\text{SO}_2 \rightarrow \text{SO}_4$ rate based on calculated $[\text{OH}]$ and $[\text{HO}_2]$ from photochemical model; constant heterogeneous oxidation rate applied as well
U of S/RDDM	No chemistry, total sulfur treated
UWATM-SOX	Linear $\text{SO}_2 \rightarrow \text{SO}_4$, diurnally varying rate

* See table 1 for model reference and model type.

distance greater than 50 km has been reported by van den Hout and van Dop (1981). They describe general features of the models and the various formulations of the physical and chemical processes. Twenty-nine models were reviewed. A summary of the models, with their individual characteristics, is given in Table 4. A comparison of model characteristics as given in Table 3 compared to those given in Table 4 will show some apparent discrepancies for the same models that appear in both tables. Reasons for these differences probably result from a lack of "hands-on" experience with each model by the authors who prepared the tables. Model developers may disagree with how their model has been characterized. Therefore, the tables are presented as the authors included them in their reports without interpretation by the sub-group report authors.

The U.S. Department of Energy recently sponsored a mesoscale model validation workshop at the Savannah River Laboratory. The proceedings of this workshop was published by the Savannah River Laboratory (1981). The participants of the workshop included representatives from nine laboratories funded by the Department of Energy. A common emissions source-term, meteorological data, and observed krypton-85 concentrations were used to evaluate model accuracy for each of the models. The model calculations were compared to the observed Kr-85 concentrations collected at 13 stations located at distances as far away as 150 km. This study is of particular importance to the Local/Mesoscale Subgroup of the U.S./Canada Transboundary Committee because of the distance for which predictions were compared with observations.

A statistical evaluation of the models showed a degradation in the ability to predict Kr-85 concentrations, as the averaging time for calculations and observations was reduced. The predictions for annual periods were reasonably accurate with the weighted-average squared correlation coefficient (R^2) for all models equal to 0.74 for annual, 0.28 for monthly, 0.21 for weekly, and 0.18 for twice-daily predictions. Model performance varied considerably within each category, however, and it was concluded that "the more complex, three-dimensional models provide only marginal increases in accuracy." A general conclusion of the evaluation was that the overriding factor in the calculation accuracy is the accurate description of the wind field and that this is more important than further improvements of the numerical accuracy of complex models.

TABLE 4. SUMMARY OF MODELS ACCORDING TO THE CLASSIFICATION GIVEN IN SECTION III (van den Hout and van Dop, 1981)

Model	Pollutant ¹	Scale ²	Type ³	Mathematical framework ⁴	Averaging time ⁵	Costs of operation ⁶
ARL	1	s/m	c/d	h	e	l
ASTRAP	2	s	c/d	h	l	l
BNL	2	s	c/d	h	l	l
Bolin and P.	1	s	c/d	h	l	l
Carmichael/P.	3	s	c/d	e	e	h
CEGB (SS)	1	m	c	a	e/l	l
CEGB (TD)	2	s	c	l	e/l	l
CIT	3	m	c	e	e	h
EPA	2	s	c/d	l	e	l
EPRI/ERT	3	s	c/d	e	e	h
EURMAP/ENAMAP	2	s	c/d	l	l	m
Fisher	2	s	d	a	l	l
Gillani	2	m	c	l	e	l
KNMI	3	m	c/d	e	e	h
Liraq	3	m	c	e	e	m
Mesoplume	2	s/m	c/d	l	e	l
Mesopuff	2	s/m	c/d	l	e	l
Mesogrid	2	s	c/d	e	e	m
Mesos	4	s	dos	l	e	l
OECD	2	s	c/d	l	e/l	m
PNL	2	s	c/d	l	l	l
Pseudo Spectral	2	s	c/d	e	e/l	l
SAI	3	m	c	e	e	h
SAI/EPA	2	s	c/d	e	e	m
Tald	4	s	dos	l	l	l
TDMB	1	m	c	e	e	m
Teknetron	2	s	c/d	e	e	l
TNO	2	m	c	a	l	l
Veltischeva	2	s	c/d	e	e	m

¹ 1 = inert pollutants, 2 = chemically reactive pollutants modeled with conversion rates or decay constants, 3 = various pollutants including (photo)chemical reactions, 4 = radioactive pollutants.

² s = synoptic, m = mesoscale.

³ c = determines ambient concentrations, d = deposition oriented, dos = dosage oriented.

⁴ a = analytic, e = eulerian, h = hybrid, l = lagrangian.

⁵ e = episodic, l = long term.

⁶ l = low, m = medium, h = high.

4 SUMMARY AND RECOMMENDATIONS

4.1 Concentrations and Deposition on a Local Scale

Although the emphasis in transboundary pollution has been for travel distances of the order of 1000 km or more, the understanding and prediction of atmospheric processes occurring in the local to mesoscale range (up to several hundred kilometers from the source) are also important. Concentrations and deposition from sources within local and mesoscale distances may augment or even dominate those from long-range transport and may be a factor that has to be taken into account if we are to develop accurate regional sulfur and nitrogen budgets. Finally, apart from the aspect of mesoscale transboundary transport, improved knowledge and modeling of mesoscale effects may improve the predictions of the models that are used for long-range transboundary transport.

(a) Sulfur Compounds

There is a wide range of estimates in the literature of the importance of nearby sources to the concentration and deposition of sulfur compounds at a given point. Most studies of both point and urban sources indicate that from a few percent to about 20% of the sulfur emitted from a source is converted in the long term to sulfate and deposited within 50 or 100 km of its source. If this is the case, then in the long term most of the emitted sulfur is available for long-range transport. There is less agreement with respect to deposition of emitted sulfur during precipitation events. Some investigators have found that 10 to 20% of deposited sulfur is accounted for by nearby sources, while in other studies the local contribution may account for most of the total deposition. The fraction deposited during a precipitation event may be as important as the long-term fraction because some receptors respond on short as well as long time scales.

These apparently contradictory results may be due to data interpretation or they may be due to real factors, including strength, composition and emission height of the local sources; the degree of pollution in the incoming air mass interacting with the local emissions; and meteorological factors, not

the least of which is whether or not dry and wet deposition is occurring. If precipitation is not occurring, the rate of dry deposition by diffusion to the surface will depend upon the nature and roughness of the surface, the height at which pollutants are emitted into the atmosphere, the height to which they are mixed into the atmosphere, and the form (species) of the sulfur. The last point is important because the rate of dry deposition of sulfur dioxide is greater than that for sulfate. The more quickly the sulfur dioxide is converted to sulfate, the lesser is the fraction of emitted sulfur that will be deposited locally in the absence of precipitation. Processes that control the rate of conversion to sulfate will be described in detail in Subgroup Report 2F - A.

When cloud and rain water are present, the rate of conversion and deposition of emitted sulfur dioxide may be much more rapid than under dry conditions and, therefore, the influence of local emissions may be relatively large. The conversion of SO_2 to sulfate will depend upon the chemical makeup of the cloud and rain water including the concentration of oxidants and catalysts, and the fraction of time that the polluted air parcel spends in a cloud. The rate of removal of sulfur by precipitation will, in addition, depend upon the frequency and intensity of the precipitation.

Despite the possible variability in the relative influence of local sources, enough evidence now exists of the rapid conversion and deposition of local SO_2 emissions to warrant further research and analysis into the important factors governing deposition close to a source. Furthermore, models capable of accounting for local deposition should be applied, tested, and further improved.

(b) Nitrogen Compounds

Nitrogen oxides are transformed to nitrate more quickly than sulfur dioxide is transformed to sulfate. Furthermore, a large fraction of these nitrogen oxides are emitted close to the ground (from vehicles) greatly enhancing the potential for near-source deposition. One might expect, therefore, that the local deposition of nitrogen oxides will be more pronounced than that for sulfur dioxides. However, less information exists

about the local deposition of nitrogen compounds than about sulfur, although some investigators have estimated that the deposition of nitrate in the vicinity of St. Louis is comparable in magnitude to the emissions from that city.

(c) Oxidants

Transboundary flow of oxidants and precursor pollutants has been observed in southwestern Ontario for many years. The high ozone concentrations are believed to be associated with warm humid air moving into the area from the south and southwest directions. There is some evidence that local emissions cause increased concentrations of ozone downwind of the sources. Researchers have reported a relationship between "weather fleck" on tobacco and the bronzing of bean crops in the areas north of Lake Erie with high ozone concentrations.

(d) Organics

The Joint Air Pollution of the St. Clair-Detroit River areas by the International Joint Commission reported B(a)P concentrations in samples of suspended particulate matter collected in the Port Huron, Windsor, and Detroit areas during 1967 and 1968. The results showed elevated levels of this carcinogen in parts of Windsor and Detroit. The Windsor concentrations were similar to those reported in cities that have steel mills. Since there are no steel mills in Windsor, the suspected cause of the elevated organics concentrations is local transboundary flow from the steel mills in Wayne County, Michigan.

(e) Suspended Particulate Matter

Much of Detroit, Michigan, and Windsor, Ontario, experience unacceptably high concentrations of suspended particulate matter. The International Michigan-Ontario Air Board report for 1980 indicates that, for most of the Detroit-Windsor area, the air quality objectives for particulate matter were exceeded in spite of the fact that there have been significant decreases in annual ambient values from 1971 to 1974 and again in 1980 (from

1975 to 1979, there was no significant decrease in annual values of suspended particulate matter).

The Ontario Ministry of the Environment Annual Reports on Ambient Air Quality in Windsor, Ontario, show a high correlation between elevated concentrations of suspended particulate matter and winds from the direction of Wayne County, Michigan. It would appear, therefore, that local and mesoscale transboundary flow of suspended particulate matter occurs from Wayne County to Windsor; 90% of the emissions of particulate matter in that area come from the Wayne County area.

Alkaline particulate material introduced into the atmosphere from wind blown dust and fugitive dust from industrial or mining activities may neutralize already present acidic particles and may modify the chemical conversion processes even on the local to mesoscale. There is evidence, for example, of emissions of alkaline dust from cement plants in the vicinity of Syracuse, New York, raising the pH of precipitation sampled in their vicinity.

(f) Fluorides

There has been documented evidence of the transport of fluorides emitted by aluminum smelters located near Massena, New York, into the neighboring area of Cornwall Island, Ontario. Investigations are presently underway to determine whether or not these have caused any health effects.

4.2 Survey of Local and Mesoscale Models

The Regional Modeling Subgroup Report (2F - M) includes a detailed investigation of eight long-range transport models and their computational results. These models produce transfer matrices for the purpose of determining source-receptor relationships over eastern North America. The transfer matrices from the long-range models indicate that, at the nine sensitive receptor points, from 10 to 40% of the annual wet sulfur deposition comes from sources within 300 km while the remaining 60 to 90% comes from sources that are farther away. There are variations among the different models about the contribution from sources within 300 km and at the present

time we are uncertain about the fraction of emitted sulfur that is deposited within the emission area itself. Possible reasons for these differences are: (1) the inadequate spatial resolution to accommodate near-source region contributions to air pollution concentrations; (2) the variations in the distributions of the emissions used in the models; (3) the variations in the vertical resolution and treatment of vertical diffusion among the long-range transport models; (4) lack of detailed chemistry in the models especially close to sources; and (5) the variations in the way that the models treat the deposition processes. Therefore, consideration should be given to using local and mesoscale models for estimates at distances smaller than 300 km.

Recent modeling survey papers, which are referenced in this report, indicate that there are many models presently in use for predicting local/mesoscale air pollution concentration distributions.

(a) Local Models (transport of less than 50 km)

The dilution of pollutants emitted from sources and transported on the local scale is strongly dependent upon transport and diffusion processes, and less dependent upon chemical transformation, dry deposition, and wet deposition. Therefore, the majority of the models included in the survey do not include these processes, as they have in the past been used primarily for assessing ambient concentrations rather than deposition.

The local models may be classified into three main categories: 1) analytical, 2) Eulerian, and 3) Lagrangian. Presently, the analytical models are the most commonly used and most widely accepted for regulatory use, although they are the least versatile for incorporating the deposition processes. The Eulerian models are the most versatile but they are more expensive and complicated to use, and have not been applied as extensively on the local scale. Few decision makers are familiar with these models and able to interpret the results effectively. Also, a very small number of evaluation studies have been carried out on these models. Lagrangian models have not been used as extensively as the analytical models but they have been applied more than Eulerian models. These models are rather versatile and can include, with some simplifications, parameterizations of the deposition processes.

While more evaluation studies have been conducted on these models than on Eulerian models, the need for more extensive evaluations is apparent.

Application of all three types of models in regions with complex topography can be expected to provide results that have a greater uncertainty than those applied in flat terrain. Research is needed to determine the local performance of models in various regions (shoreline, plain, mountain valley, etc.) that incorporate deposition processes.

(b) Mesoscale Models (transport of 50 to 300 km)

The survey of dispersion models, applicable for this intermediate range, indicates the availability of more than 50 models. The survey includes the following categories of models: 1) analytical, 2) Eulerian, 3) Lagrangian, and 4) hybrid (a combination of Eulerian and Lagrangian). In addition to the more common attributes of long-range transport models, that is, transport and diffusion, physical and chemical transformation, and wet and dry deposition, other features important for intermediate-range transport are considered in the survey. These other significant features include the models' capability to: (1) resolve vertical pollutant distributions, (2) accommodate urban emissions, and (3) simulate detailed atmospheric chemistry.

A mesoscale model validation workshop was recently conducted at the Savannah River Laboratory. A report indicated that model predictions were reasonably accurate for annual averages, but that the accuracy decreased with decreasing averaging times. A general conclusion from this workshop was that the overriding meteorological factor in improving computational accuracy was the accurate description of the wind field. The model evaluations were performed using a non-reactive pollutant, Kr^{85} . Of course, for reactive pollutants such as sulfur oxides, improvements will have to be made in the chemistry modules.

4.3 Recommendations for Future Work

(1) Detailed field studies should be carried out to learn more about the concentration and deposition fields within 100 to 200 km of a major source of sulfur for an extended period to determine how much of the emitted sulfur is being deposited locally. Results should be averaged over both extended and shorter periods during specified meteorological conditions. These studies would include the measurement of concentrations in the ambient air and in precipitation, not only of sulfur compounds but, if possible, other substances that may effect the sulfur chemistry, such as oxides of nitrogen, oxidants, and catalysts such as trace metals. These measurements should be carried out with sufficient spatial extent and resolution to shed insight into the transformation rates of the various chemical species and their budgets.

(2) A careful examination should be made of the existing data on local and mesoscale deposition that can be found in the literature in order that maximum use be made of them.

(3) Existing, less complex, analytical models may be appropriate for use out to distances of a few kilometers. Existing, more complex, Lagrangian, Eulerian, or hybrid models may also be appropriate for distance scales of the order of a few tens to a few hundred kilometers. The local models need to be improved to better account for chemical transformation, and wet and dry deposition processes.

(4) Mesoscale models that are used for large spatial scales with corresponding time scales of the order of several hours to a few days must include chemical transformation, wet and dry deposition processes, and transport and diffusion processes with suitable vertical resolution.

(5) As indicated in the survey, there are numerous models in all of the above categories. It is difficult, however, to select a model, or combination of models, for use in a particular application. With so many models available, which appear to have been developed on sound theoretical principles, the Work Group recommends that research and development of new models be given a lower priority than evaluating the performance of existing

models. The models that the Work Group feels are most likely to be suitable for application in the local/mesoscale range, which include the important physical and chemical processes and complement the long-range transport models, are the hybrid models.

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