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A SENSITIVITY ANALYSIS AND PRELIMINARY EVALUATION OF RELMAP INVOLVING FINE AND COAPSE PARTICULATE MATTER

A SENSITIVITY ANALYSIS AND PRELIMINARY EVALUATION OF RELMAP INVOLVING FINE AND COARSE PARTICULATE MATTER

by

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AFFILIATION

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ABSTRACT

In response to the new, size discriminate federal standards for Inhalable Particule Matter, the REgional Lagrangian Model of Air Pollution (RELMAP) has been modified to include simple, linear parameterizations which simulate the chemical and physical processes of fine and coarse particulate matter.

Because these new, simplified parameters are only accurate to a limited degree, they may be upgraded or replaced in the future with more sophisticated parameters as further research is conducted. As an initial step in this possible refinement, RELMAP has been subjected to a sensitivity analysis in which the effect of inducing a +/- 50% change in the three major parameterizations (transformation rate and wet and dry deposition rates) involving the simulation of fine and coarse particulate matter has been examined. Simulated concentrations of fine and coarse particulate matter proved to be most sensitive to the wet deposition of fine and coarse particulate matter, respectively; fine concentrations were somewhat sensitive to the transformation rate of sulfur dioxide (SO_2) into sulfate ($SO_4^=$), and less sensitive to the wet deposition of SO_2, and the dry deposition of fine particulate matter and SO_2; and finally coarse particulate concentrations were somewhat sensitive to the dry deposition of coarse particulate matter.

In order to assess the model's abilities, and to determine just how accurately these new parameters simulate the actual physical and chemical processes of the atmosphere, RELMAP was evaluated for the summer of 1980, using emissions data from the NAPAP Version 5.0 emissions inventory, monitoring data from the Inhalable Particulate Network and meteorological data from the National Climatic Data Center. Unfortunately, several obstacles limited the scope of this evaluation; the two most important being the omission of open source emissions from the NAPAP inventory, and the spatial and temporal incompatibility of the IPN data. Given the nature of these deficiencies, it is not surprising that RELMAP significantly underpredicted the concentrations of fine and coarse particulate matter. The model did, however, exhibit some skill in its simulation of the concentrations, producing correlation coefficients of 0.53 and 0.33 for fine and coarse particulate matter, respectively.

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LIST OF ACRONYMS AND SYMBOLS

Acronyms

EMSL	-	Environmental Monitoring Systems Laboratory
ENAMAP	-	Eastern North American Model of Air Pollution
EURMAP	-	EUropean Model of Air Pollution
IP	-	Inhalable Particulate
NAPAP	-	National Acid Precipitation Assessment Program
NAAQS	-	National Ambient Air Quality Standards
OAQPS	-	Office of Air Quality Planning and Standards
RELMAP	-	REgional Lagrangian Model of Air Pollution
SCC	-	Source Classification Code
SSI	-	Size Selective Inlet
TSP	-	Total Suspended Particulate

Symbols

a	-	empirical factor used to calculate wet deposition
b	-	empirical exponent used to calculate wet deposition
CO	-	carbon monoxide
HCl	-	hydrogen chloride
HF	-	hydrogen fluoride
k	-	von Karman constant (0.4)
K_d	-	dry deposition rates
K_t	-	transformation rates
K_w	-	wet deposition rates
L	-	Monin-Obukhov length (cm)
M	-	mass
NH_3	-	ammonia
NO_x	-	nitrogen oxides
Pb	-	lead
R	-	rainfall rate used to calculate wet deposition (mm/h)
r_p	-	surface resistance to deposition (1.0 s/cm)
S	-	Stability category
SO_2	-	Sulfur dioxide
$SO_4^=$	-	Sulfate
u_*	-	friction velocity (cm/s)
v_d	-	dry deposition velocity (cm/s)
VOC	-	volatile organic carbon
Y_c	-	stability factor
z	-	height above surface (cm)
z_o	-	surface roughness scaling length (cm)

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SECTION 1

INTRODUCTION

The primary National Ambient Air Quality Standard (NAAQS) for particulate matter was established in 1970 with the enactment of the Clean Air Act. The values of the standard were based upon state-of-the-art information concerning the health effects of ambient concentrations of Total Suspended Particulate (TSP) matter and other environmental factors. In 1977, the Clean Air Requirement Act called for a reappraisal of this NAAQS. One reason for this reappraisal was a shift in emphasis from TSP, which ranged in size from 0.0 to 50.0 um, to smaller, size discriminate Inhalable Particulate (IP) matter, which ranged in size from 0.0 to 15.0 um. The IP was comprised of fine particulate matter (FINE-10), which included particles less than 2.5 um in diameter, and coarse particulate matter (COARSE-15), which initially included particles greater than or equal to 2.5, but less than or equal to 15.0 um.

Emphasis was placed on the smaller size particles for two reasons. First, additional research into the health effects of particulate matter revealed that the smaller particles were not only able to penetrate deeper into the respiratory system, but their expulsion rate was also lower than the larger particles. Secondly, naturally occurring dust particles with diameters

greater than 10.0 - 15.0 um often made up a large percentage of the TSP mass collected by high volume samplers. This large natural contribution, when combined with anthropogenic sources, often resulted in TSP concentrations exceeding either the 75.0 ug/m^3 annual geometric mean, or the 260.0 ug/m^3 daily mean (Hinton et al., 1984).

In 1981, after reviewing EPA's Clean Air Science Advisory Committee's recommendation and the concurrent International Standards Organization Task Group recommendations, the Office of Air Quality Planning and Standards (OAQPS) decided that the revised standard for ambient air concentrations of IP should be based upon a 10 um rather than a 15 um criteria (Hinton et al., 1984). Therefore, COARSE-15 was replaced by COARSE-10, which included particles greater than or equal to 2.5 um, but less than or equal to 10.0 um. The proposed new standard would allow ambient air concentrations of IP to reach an annual arithmetic average between 50 and 65 ug/m^3 and a daily maximum between 150.0 and 250.0 ug/m^3 (Federal Register, 1984).

As a result of the revised NAAQS standards for ambient air concentrations of primary particulate matter, OAQPS has expressed the need for size discriminate particulate models in order to assist in regulatory planning. Shifting the emphasis onto the smaller particles increases the importance of regional scale models. Much of the mass of the smaller particles results from gas to aerosol conversion which is a slow process that occurs over regional spatial scales as opposed to urban scales. Therefore, in response to the promulgation of the new size discriminate federal standards for IP, the REgional Lagrangian

Model of Air Pollution (RELMAP) has been modified to include simple, linear parameterizations which simulate the chemical and physical processes of FINE-10 (including the conversion of SO_2 to $SO_4^=$) and COARSE-10. Because the contribution of nitrogen chemistry in the formation of particulate matter is thought to be negligible at this time, it is ignored by the model. It is important to remember that this modified version of RELMAP represents an initial step in the linear, regional modeling of particulate matter and therefore must be looked upon as an interim model.

The origin of RELMAP dates back to the mid-1970's, when SRI International developed a Lagrangian puff air pollution model called the EUropean Model of Air Pollution (EURMAP) for the Federal Environment Office of the Federal Republic of Germany (Johnson et al., (1978). This original version of the model only simulated monthly concentrations and wet and dry depositions of SO_2 and $SO_4^=$, for thirteen countries in central and western Europe. During the late 1970's, the U.S. EPA sponsored SRI International to modify EURMAP so that it could be applied to eastern North America. This modified version of the model, now called the Eastern North American Model of Air Pollution (ENAMAP) was also capable of simulating monthly concentrations as well as wet and dry depositions of SO_2 and $SO_4^=$ (Bhumralkar et al., 1980; Johnson, 1983).

During the early and middle 1980's, EPA continued to modify and improve the model to increase its flexibility and its scientific credibility. Now, at the request of OAQPS, simple

parameterizations involving fine and coarse particulate matter have been incorporated into the model.

This report examines the incorporation of these new parameterizations, first through an abbreviated discussion of their theoretical aspects as presented in Chapter 2. For a more in-depth discussion of these parameterizations, the reader is referred to the RELMAP User's Guide (Eder et al., 1986). In order to determine the sensitivity of the model to these parameterizations, a sensitivity analysis of RELMAP has been provided in Chapter 3. Chapter 4 provides a preliminary model performance evaluation to help assess the model's abilities and to determine how accurately the parameters simulate the actual physical and chemical processes of the atmosphere. Conclusions and recommendations for future work are provided in Chapter 5.

SECTION 2

MODEL BACKGROUND

RELMAP is a mass-conserving, regional-scale Lagrangian model that performs simulations over 1^{o} by 1^{o} grid cells covering the eastern two-thirds of the United States and southeastern Canada as depicted in Figure 2.1. The north-south and east-west boundaries of the model's domain extend from 25^{o} N to 55^{o} N latitude and from 60^{o} W to 105^{o} W longitude. Discrete puffs of SO_2, $SO_4^{=}$, fine and coarse particulate matter are released at twelve hour intervals from each of the 1350 grid cells that contain sources. As illustrated in Figure 2.2, the puffs are then subjected to linear chemical transformation and wet and dry deposition processes as they are transported across the model's domain. The suspended mass and deposition for each puff is apportioned into the appropriate grid cell based upon the percentage of puff over that grid cell. The rate of change in the pollutant mass resulting from the transformation and wet and dry deposition process is directly proportional to the total mass and is defined through the following linear equations:

SO_2:

$$\frac{dM_1}{dt} = -M_1(K_t + K_{d1} + K_{w1}); \qquad (2.1)$$

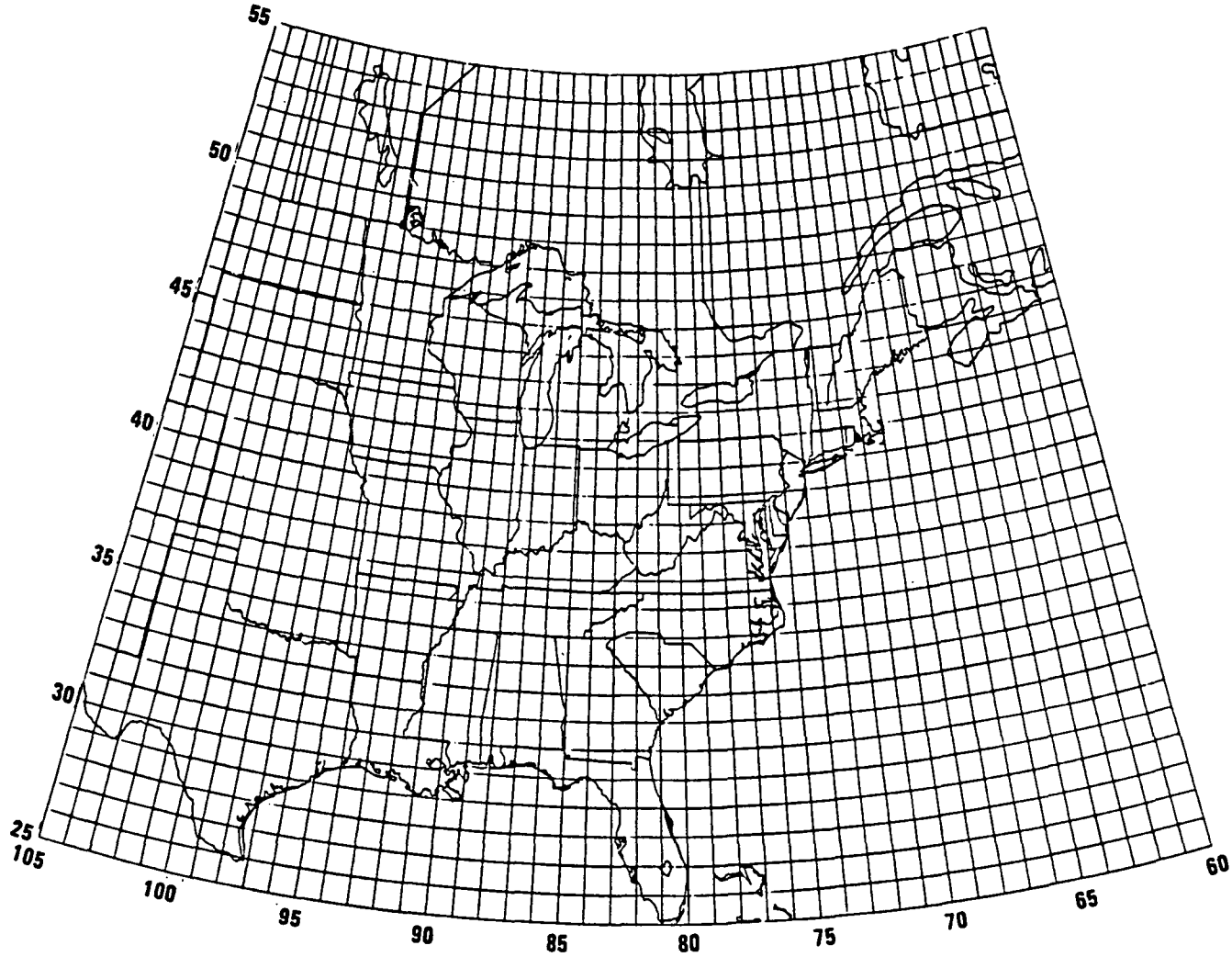


Figure 2.1 RELMAP's 45° x 30° longitude-latitude domain.

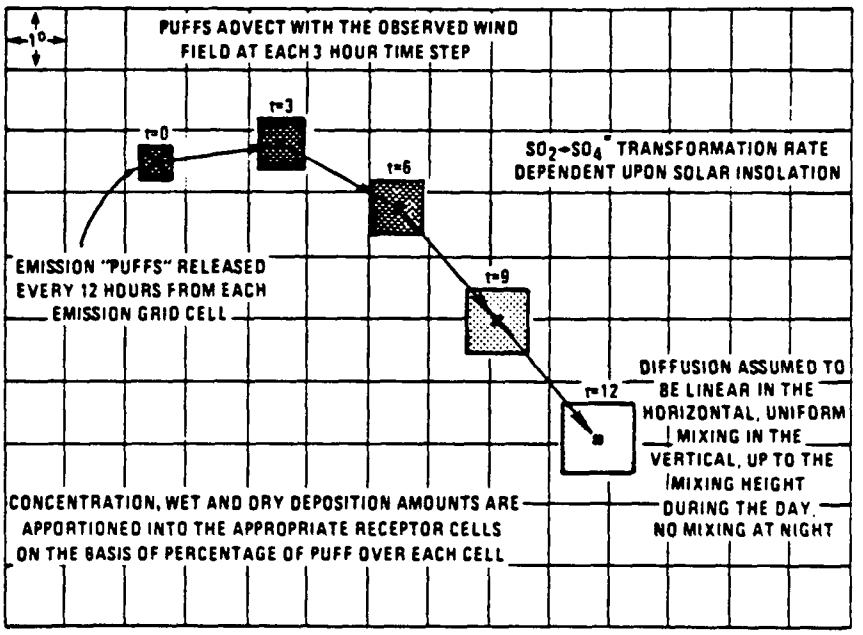


Figure 2.2 Depiction of RELMAP parameterizations.

$SO_4^=$: $$\frac{dM_2}{dt} = -M_2(-\tfrac{3}{2}K_t + K_{d2} + K_{w2}); \quad (2.2)$$

Fine Particulate: $$\frac{dM_3}{dt} = -M_3(K_{d3} + K_{w3}); \quad (2.3)$$

Coarse Particulate: $$\frac{dM_4}{dt} = -M_4(K_{d4} + K_{w4}); \quad (2.4)$$

where M_i is the mass of the respective pollutants (expressed in ktons), t is the time (h), and K_t is the transformation rate of SO_2 into $SO_4^=$, K_{di} is the dry deposition rate and K_{wi} is the wet

deposition rate, of the respective pollutants. The 3/2 factor used in Equation 2.2 represents the ratio of molecular weights between $SO_4^=$ and SO_2.

Transport and Diffusion

Dispersion generated by small-scale turbulence is not nearly as significant as long term transport and deposition processes for regional-scale models such as RELMAP. Because of this, the model simulates both horizontal and vertical diffusion through simple parameterizations. RELMAP divides the atmospheric boundary layer into three layers as seen in Figure 2.3. The first layer is between the surface and 200 m, while the second layer is between 200 and 700 m. The depth of the third layer is variable, depending upon the seasonal-mean maximum mixing height, and is assumed to be 1150 m during the winter, 1300 m during the spring and fall, and 1450 m during the summer (Endlich et al., 1983).

During the unstable regimes of midday periods, pollutants from both area and point sources become well mixed up to the mixing height long before they are transported a distance equal to the spatial resolution of the grid. Therefore, it is assumed that instantaneous and complete mixing occurs within the three layers of the model during the unstable daylight hours. However, after sunset, when mixing is prohibited by stable conditions, point and area source emissions are confined to the separate

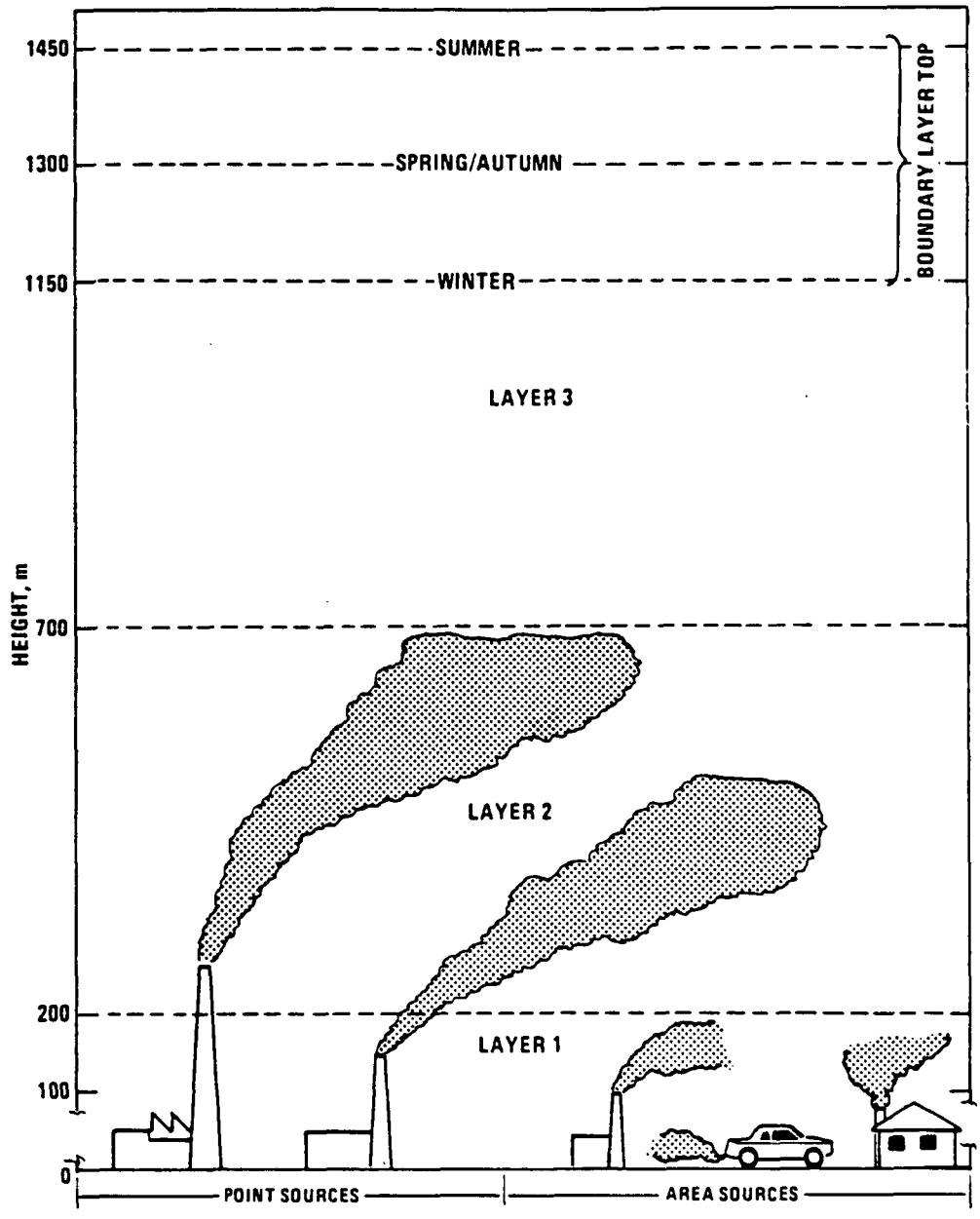


Figure 2.3 Three layer vertical profile with nighttime allocation of emissions.

layers into which they are emitted. As again illustrated in Figure 2.3, all area source emissions remain in Layer 1, within 200 m of the surface, while emissions from point sources are allocated into Layer 2, accounting for typical plume rise, which averages several hundred meters (Briggs, 1984).

RELMAP assumes that horizontal diffusion of the puffs occurs

at a constant rate so that the size of the puff increases at a rate of 339 km^2/h, and that the distribution of the mass of pollutant in the puff remains homogeneous at all times. The puff expansion rate is based upon work by Pack et al. (1978), who performed calculations on long range trajectories. Each of the puffs is transported using vertically weighted and horizontally and temporally interpolated wind fields until the puff is either transported off the grid, or the amount of mass in the puff falls below a predetermined minimum value. The puff remains an indivisible entity. Vertical shear of the puff is not directly considered as the mass of pollutant in each of the three layers is transported in the same direction and at the same speed. The transport velocity of the puffs is determined by integrating the mass-weighted u and v components of the three layers, which are derived from the wind velocities from the grid cell containing the centroid of the puff. Surface winds are used in the lowest layer, 850 mb winds are used in the third layer, and a weighted average of (0.2) surface and (0.8) 850 mb wind velocities are used for the second layer.

Transformation

RELMAP treats fine and coarse particulate matter as independent non-evolving pollutants, that is physical and/or chemical transformation of fine particulate matter to coarse particulate matter are considered negligible. This premise is

supported by particle size distributions obtained from monitoring data (Suggs et al., 1981). As seen in Figure 2.4 the size distribution of particulate matter generally indicates a bimodal distribution with peaks in the fine and coarse particle size ranges and a deep oscillating gap between 1 and 5 um.

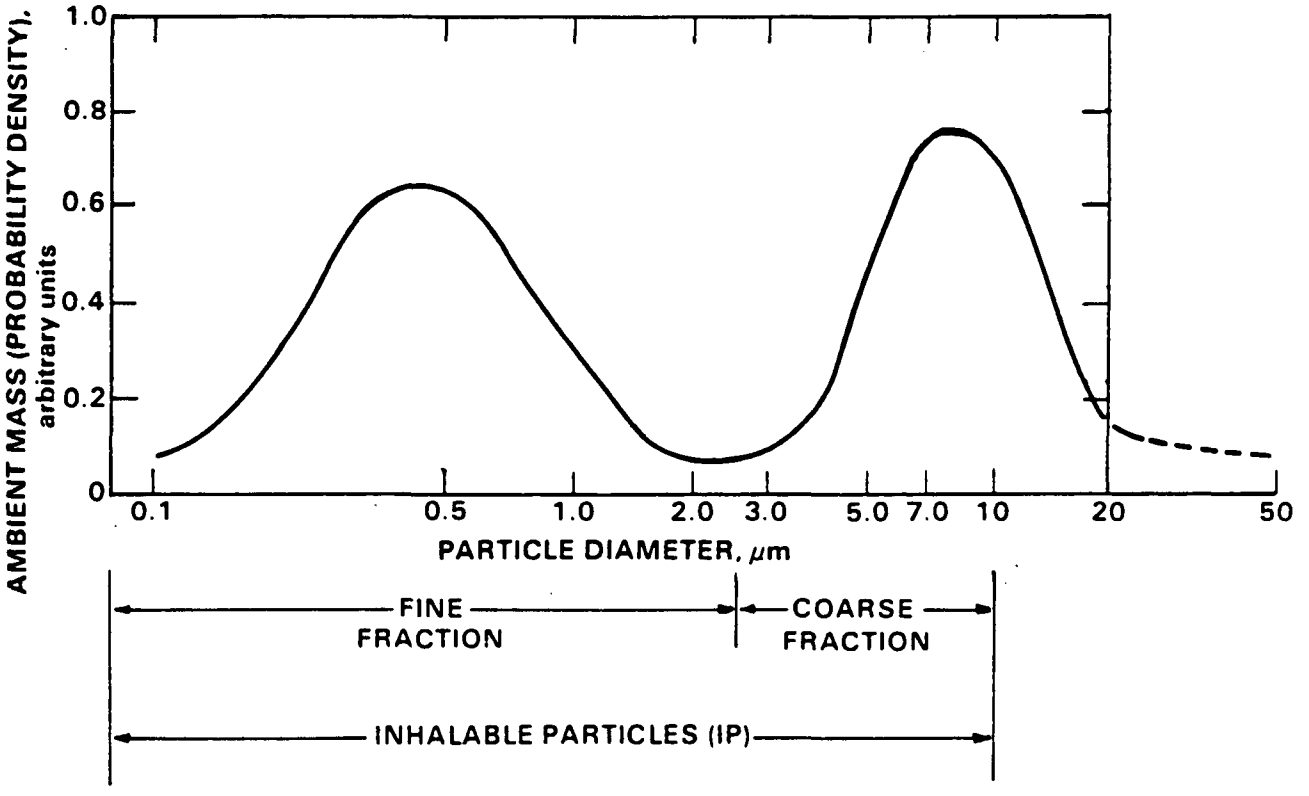


Figure 2.4 Bimodal probability distribution of particle size.

RELMAP does, however, consider the transformation of SO_2 into $SO_4^=$. This oxidation process varies primarily with solar insolation (i. e. diurnally, latitudinally, and seasonally) and with moisture content. The model simulates this process through the use of a heterogeneous component and a homogeneous component. The heterogeneous component accounts for the more rapid transformation that occurs under saturated conditions, while the

homogeneous component simulates transformation that occurs under dry conditions.

Seasonal variation in the transformation rates were examined by Meagher et al., (1983) who determined that average morning rates ranged from a low of 0.15%/h during the winter to a high of 1.30%/h during the summer. In a similar study, Altshuller (1979) concluded that noontime winter transformation rates averaged about five times less than noontime summer transformation rates.

Diurnal fluctuations were found to be much larger during the summer months than during the winter months. Husar et al., (1978) found summer diurnal transformation rates that ranged from a minimum of 0.5%/h during the night to a maximum of between 2.0 to 8.0%/h at solar noon. Conversely, Meagher and Olszyna (1985) could only detect slight diurnal variation in the transformation rates during the winter months.

More recent field studies have indicated that in-cloud processes are also very important in the transformation of pollutants. The rate of transformation can be increased by an order of magnitude in saturated conditions, depending upon the cloud height, precipitation efficiency and mass of SO_2 in the mixed layer (Isaac et al., 1983). Based upon a theoretical algorithm developed by Scott (1982), the magnitude of this heterogeneous component was set to 7.0%/h during the winter, 11.0%/h during the spring and fall, and 15.0%/h during the summer.

Figures 2.5 and 2.6 illustrate the relationship between the composite transformation rate (heterogeneous and homogeneous

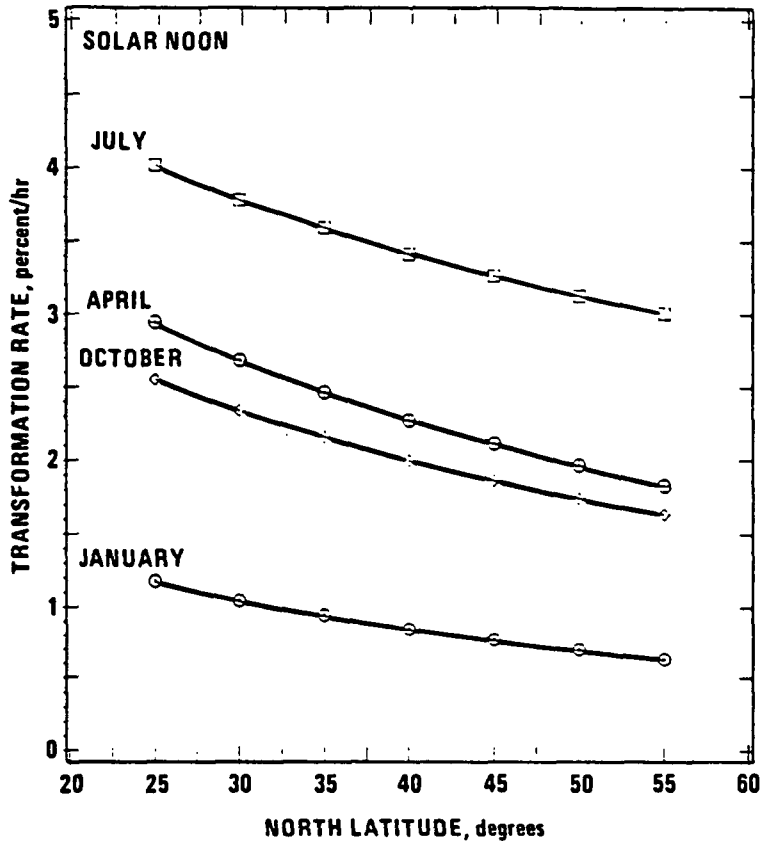


Figure 2.5. Latitudinal variation in the composite transformation rate of SO_2 to $SO_4^=$.

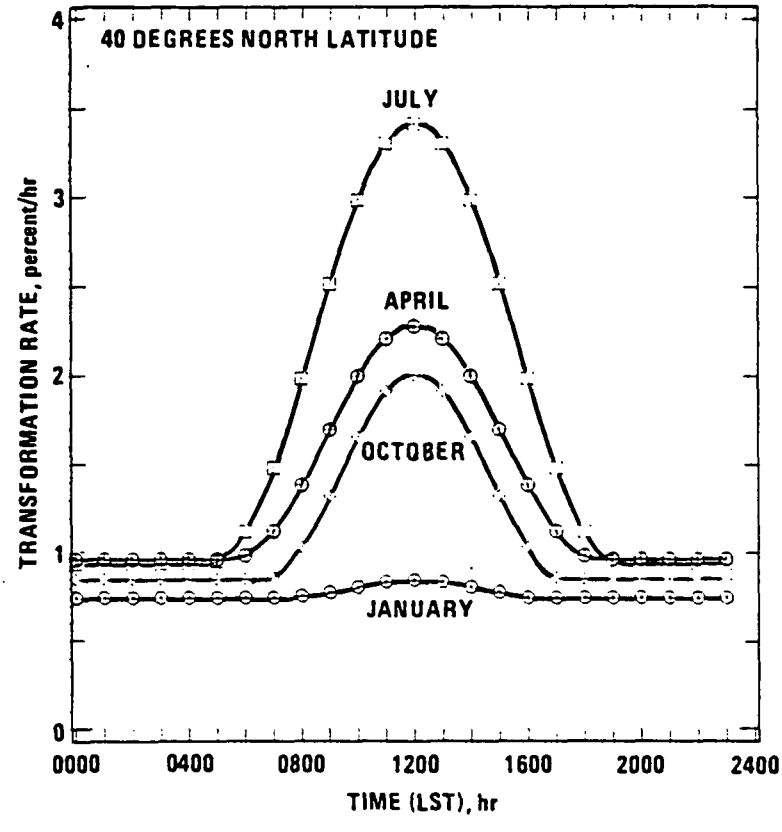


Figure 2.6. Diurnal variation in the composite transformation rate of SO_2 to $SO_4^=$.

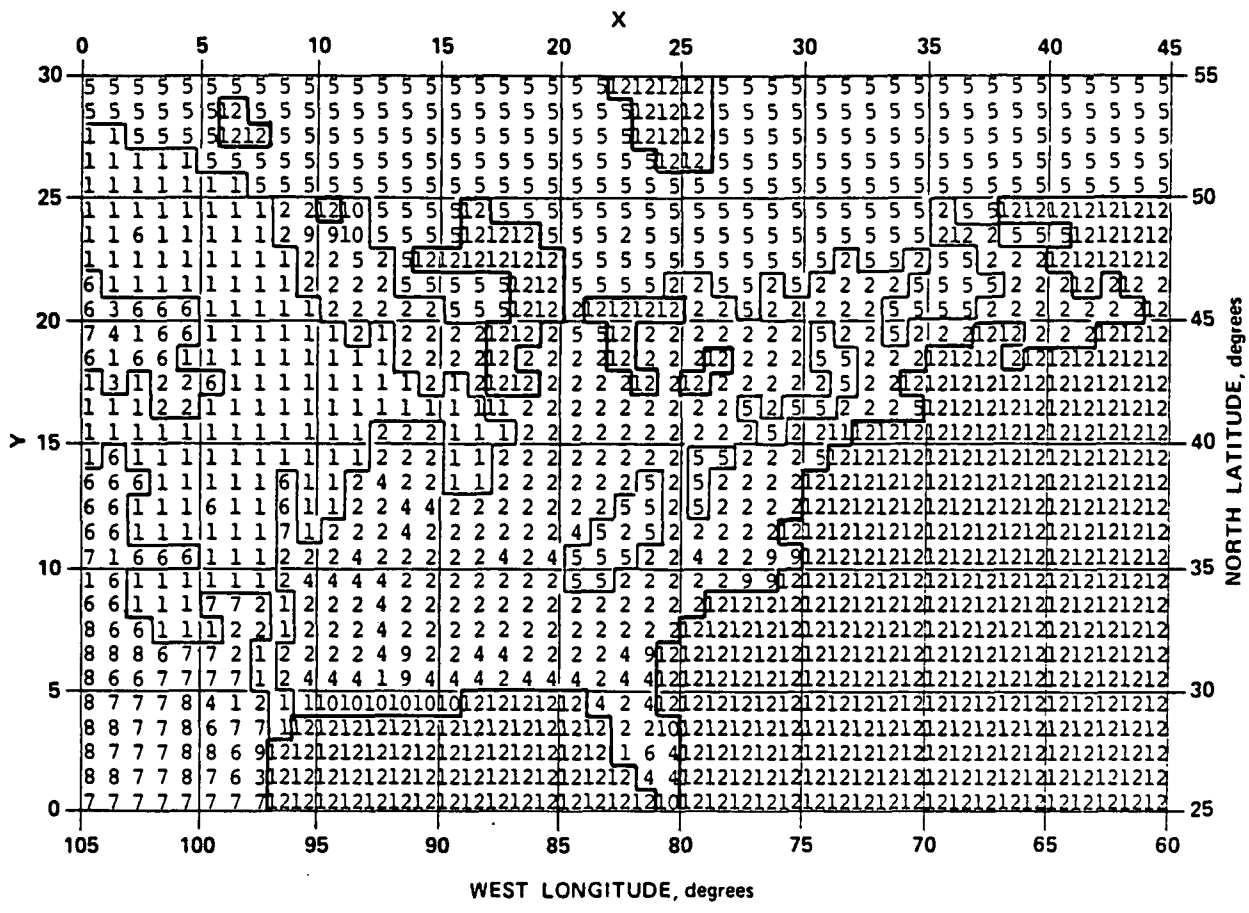
components) and the time of day, season and the latitude as calculated in RELMAP.

From Figure 2.5 it is evident that at solar noon, the composite transformation rate incorporated into the model is highest during July (approximately 4.0%/h at 25° N and 3.0%/h at 55° N) and lowest during January (1.2% at 25° N and 0.6%/h at 55°). Figure 2.6 illustrates that the diurnal variation exhibited by the composite transformation rate at 40° N is also greater during July (0.9%/h at midnight LST and 3.4%/h at solar noon) than during January (0.7%/h and 0.8%/h).

Dry Deposition

Dry deposition of SO_2, $SO_4^=$, fine and coarse particulate matter is a highly variable, complex process that is parameterized in RELMAP as a function of land use, season, and stability index. Twelve land use categories, categorized by surface characteristics and vegetation type (Sheih et al., 1979), were gridded to RELMAP's 1° by 1° domain. Figure 2.7 illustrates the grid of homogeneous land use types and provides a Table listing their corresponding surface roughness scale lengths (z_o).

Dry deposition velocities (v_d), which represent the downward surface flux divided by the local concentration, were calculated as a function of land use type, stability class and season for SO_2, $SO_4^=$, and fine and coarse particulate matter. The stability classes used to determine the dry deposition velocities are the



Symbol	Land Use Type	z_0 (cm)
1	Cropland and Pasture	20
2	Cropland, Woodland, and Grazing	30
3	Irrigated Crops	5
4	Grazed Forest and Woodland	90
5	Ungrazed Forest and Woodland	100
6	Subhumid Grassland and Semiarid Grazing	10
7	Open Woodland, Grazed	20
8	Desert Shrubland	30
9	Swamp	20
10	Marshland	50
11	Metropolitan City	100
12	Lake or Ocean	0.01

Figure 2.7. Land use categories used for dry deposition calculations and corresponding surface roughness lengths (Sheih et al., 1979).

six Pasquill-Gifford categories: (A) very unstable, (B) moderately unstable, (C) slightly unstable, (D) near neutral, (E) moderately stable, and (F) very stable (Gifford, 1976). The dry deposition velocities, measured in centimeters/second, are used in the model to determine dry deposition rates.

Determination of the dry deposition velocities for SO_2, $SO_4^=$ and fine particulate matter were based upon the work of Sheih et al., (1979) and are discussed below. Coarse particulate matter dry deposition velocities, which are based upon the work of Sehmel (1980) and therefore parameterized somewhat differently, are also presented below.

The algorithm developed by Sheih et al., (1979) was modified to calculate dry deposition velocities. The parameterization used in RELMAP for the deposition of SO_2, $SO_4^=$, and fine particulate matter is given by the following:

$$V_d = ku_* (\ln (z/zo) + ku_* r_p - Y_c)^{-1} \qquad (2.5)$$

where k is the von Karman constant (0.4), u_* is the friction velocity (cm/s), z_o is the surface roughness scale length (cm) derived from the twelve land use categories, r_p is the surface resistance to particle deposition (1.0 s/cm) and Y_c is a stability factor. Details on the formulation of Equation 2.5 can be found in Sheih et al. (1979).

More recent studies (Wesely and Shannon, 1984), which are based upon micrometeorological field experiments, have determined that dry deposition of $SO_4^=$ calculated by Equation 2.5 was too

high by a factor of two. To alleviate this overestimation, the dry deposition velocities for $SO_4^=$ and fine particulate matter as calculated from Equation 2.5 were reduced by half. Typical dry deposition velocities resulting from the calculation range between 0.05 and 1.15 cm/s for SO_2, and between 0.05 and 0.50 cm/s for $SO_4^=$ and fine particulate matter, depending upon the season, the stability and the land use category.

When considering diurnal variations, use of the equations derived above is not always recommended. In order to compensate for the high nocturnal atmospheric resistance, when plant absorption is minimal, the model assumes that dry deposition velocities are reduced to 0.07 cm/s for SO_2, $SO_4^=$ and fine particulate matter, as recommended by Sheih et al. (1979).

Dry deposition velocities of coarse particulate matter are parameterized through a very similar approach in order to maintain consistency within the structure of the model. Using the same land use categories as described earlier, the model incorporates the work of Sehmel (1980) who presented plots of dry deposition velocities of particulate matter as a function u_*, z_o, particle density, and diameter. The following equation was used to determine values of u_*, which is a function of stability, wind speed and z_o:

$$u_* = ku(\ln z/zo) - Y_m)^{-1} \qquad (2.6)$$

The stability function, Y_m, was determined by using the appropriate relationships between the Monin-Obukhov length (L), surface wind speed (u) and stability class, as suggested by

Sheih et al. (1979). Determination of u_* allows the selection of an appropriate Sehmel diagram, from which the dry deposition velocity can be obtained for a given z_o. Based upon the work of Mamane and Noll (1985), who analyzed rural particulate matter characteristics, a constant particle density of 4.0 g/cm^3 was used in the equation. Unfortunately, Sehmel's study was limited to surface roughnesses less than 10 cm, while most of the land use categories used in the model had surface roughnesses greater than 10 cm, therefore it was often necessary to extrapolate the appropriate dry deposition velocity.

Unlike SO_2, $SO_4^=$, and fine particulate matter, the dry deposition velocities of coarse particulate matter are much less dependent upon the time of day and the season, therefore, diurnal and seasonal variations are considered by the model to be negligible.

Wet Deposition

The complex process of wet deposition of SO_2, $SO_4^=$, and fine and coarse particulate matter is thought to be a function of cloud chemistry, cloud type, pollutant concentration and precipitation type and rate. RELMAP, however, parameterizes this process quite simply, treating it only as a function of precipitation rate and cloud type. The wet deposition rates are based upon the work of Scott (1978), who presented graphs of washout ratios between $SO_4^=$ concentration in precipitation and $SO_4^=$ concentrations in the air. These ratios are solely

dependent upon the precipitation rate and the cloud type, where the three cloud types considered are Bergeron or cold-type clouds, maritime or warm-type clouds, and convective-type clouds. The model assumes that all winter precipitation results from the Bergeron process, that spring and summer precipitation result from the convective-type clouds, and that autumn precipitation is confined to warm-type clouds. The algorithm derived from Scott's work, which was expanded by SRI (1982) to include SO_2, is presented below:

$$\text{Wet Deposition Rate} = a R^{b} ; \qquad (2.7)$$

where a and b are seasonal empirical constants derived from the inherent relationship between the washout ratio and the precipitation rate R. Because so little is known about the wet removal processes of nonsulfate aerosols from the atmosphere, the model currently assumes identical deposition rates for $SO_4^{=}$, and fine and coarse particulate matter. This simplistic approach to the wet removal processes of nonsulfate particulate matter will be replaced in the future with more sophisticated parameterizations as the physics of this process become better understood.

As a participant in the International Sulfur Deposition Model Evaluation (ISDME), RELMAP was found to significantly overpredict $SO_4^{=}$ wet deposition amounts during the convective seasons of spring and summer using the algorithm discussed above (Clark et al., 1987). Predictions of $SO_4^{=}$ wet deposition during the non-convective months of winter and autumn were, however,

more in line with the observed. Further analysis has shown that, because wet deposition is such an efficient sink of particulate matter, the length of the simulation period is very critical. This is especially true during the convective months when precipitation rates can be very high for short time intervals. Therefore, in an effort to better simulate the convective type precipitation event, the time step used to calculate wet deposition amounts during the spring and summer were reduced from the nominal 3 h to 1.5 h. Increasing the temporal resolution, which has produced more favorable results, decreases the amount of wet deposition occurring for a given amount of precipitation.

Presented below in Table 2.1 are typical wet deposition rates calculated for a given precipitation amount (5mm/h) and for each season using the 1.5 h time interval for spring and summer and the 3 h time interval for winter and autumn.

Table 2.1 Typical, Seasonal Wet Deposition Rates for SO_2, $SO_4^=$, Fine and Coarse Particulate Matter for a Constant Precipitation Rate of 5mm/h.

Pollutant	Season	Empirical Constant a	Empirical Constant b	Wet Deposition Rate (% / Time step*)
SO_2	Winter	0.009	0.70	8.10
	Spring	0.036	0.53	17.72
	Summer	0.140	0.12	26.36
	Autumn	0.036	0.53	23.26
$SO_4^=$, Fine & Coarse PM	Winter	0.021	0.70	18.20
	Spring	0.091	0.27	24.31
	Summer	0.390	0.06	58.96
	Autumn	0.091	0.27	36.51

* Time Step = 1.5 h during spring and summer, 3.0 h during autumn and winter

SECTION 3

SENSITIVITY ANALYSIS

The simplified parameterizations, which were recently incorporated into the model, are designed to simulate the complex meteorological and chemical process involving fine and coarse particulate matter. Because of their simplicity, they may be upgraded or even replaced in the future with more sophisticated parameterizations as further research is undertaken. As an initial step in this possible refinement, RELMAP was subjected to a "local" sensitivity analysis. In this analysis, variations found in the model's output (concentrations of fine and coarse particulate matter) due to changes in the model's parameterizations are examined, while all the other parameters are held fixed.

The analysis, which employed actual meteorological and emissions data for July 1980, was performed using the currently accepted values for all of the input parameters. The meteorological data were obtained from the National Climatic Data Center located in Asheville, North Carolina, and included 12 hourly surface and 850 mb wind data and hourly precipitation data. Because Version 5.0 of NAPAP's 1980 Task Group B emission inventory was not available at the time of this analysis, the emissions data were obtained from the Version 4.0 inventory and

from Canada's Environmental Protection Service emissions inventory used in Phase III of the U.S./Canadian Memorandum of Intent on Transboundary Air Pollution (U.S./Canadian Memorandum of Intent, 1982).

The parameterizations examined in this sensitivity analysis included: the transformation rate of SO_2 into $SO_4^=$, the wet and dry deposition rates of SO_2, fine (including $SO_4^=$) and coarse particulate matter. SO_2 parameterizations are included in this analysis because it is a precursor to $SO_4^=$ and therefore to fine particulate matter. With each simulation, the values of the respective parameterizations were allowed to vary +/- 50% around their currently accepted or nominal values. A single value of 50% was selected for two reasons. First, choosing a single value would maintain consistency between and allow intercomparisons of each of the sensitivity tests. Secondly, the value of 50% was found to best represent the approximate lower and upper limits of the realistic changes found in all of the parameters. As a result, one would expect that the subsequent changes found in the simulations of fine and coarse particulate matter concentrations would correctly represent the range in which the actual concentrations would vary, given that the exact physics and chemistry had been incorporated into the model.

Results of the sensitivity analysis were recorded along a specific transect that stretched across the model's domain from Alabama to Quebec as seen in Figure 3.1. The fifteen grid cells that comprise the transect were chosen because they provide a good representation of the actual range of concentration values found for both fine and coarse particulate matter in North

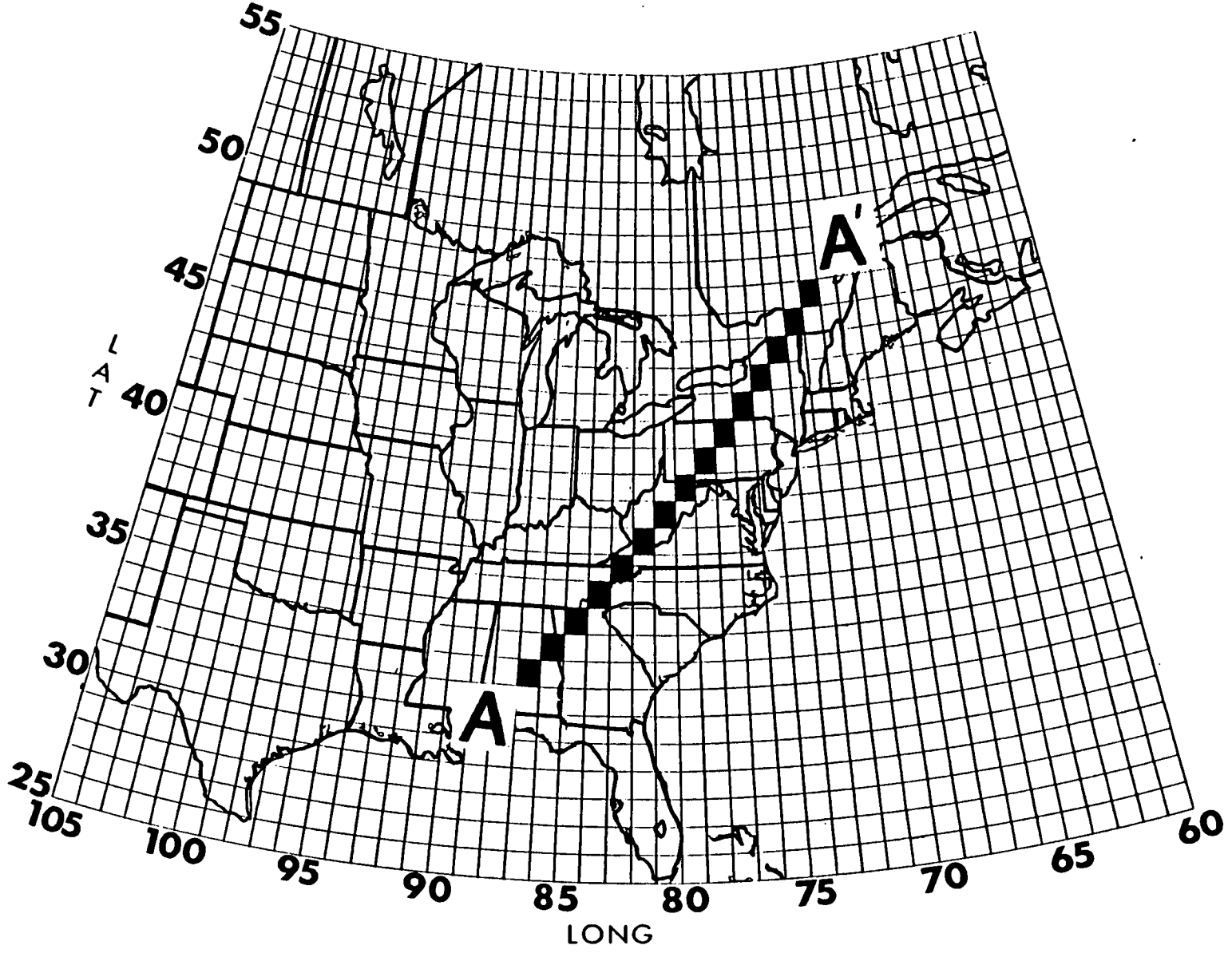


Figure 3.1 RELMAP domain with sensitivity analysis transect.

America. Results from the seven tests are presented graphically in Figures 3.2 through 3.8. Figures 3.2 and 3.3 illustrate the sensitivity of coarse particulate matter concentration to changes in the wet and dry deposition rates of coarse particulate matter, respectively. Figures 3.4 through 3.8 depict the sensitivity of fine particulate matter concentrations to changes in the transformation rate, and the wet and dry deposition rates of SO_2, and fine particulate matter. Although the model treats $SO_4^=$ and fine particulate matter as mutually exclusive pollutants, the two are combined as one in this graphical analysis and simply referred to as fine particulate matter, unless otherwise noted.

Each analysis consists of two graphs. The first graph depicts a transect of the concentration field illustrating the absolute changes that occur when a parameter is allowed to vary by +/- 50% around its nominal or base case value. The second graph of each analysis illustrates the relative, with respect to the base case, changes that occur along the transect. The abscissa for each of the plots represent the fifteen grid cells that form the transect from A to A'. It should be noted, that the scale of the ordinate, which represents either the actual concentration (expressed in ug/m^3), or the relative concentration (percent of the base case) can vary significantly from plot to plot, depending upon the specific parameter and the model's sensitivity to that parameter.

In each graph, the asterisk represents the concentration of fine and coarse particulate matter that results when the parameter being tested is reduced to 50% of its nominal value. The diamond represents the base case, where the parameter is left

at its nominal value, and the circle represents 150% of the parameter's nominal value. Caution should be exercised when examining the relative graphs at grid cells fourteen and fifteen. Concentrations at these two grids cells, which are over Ontario and Quebec, are so small that even minute changes in the magnitude of the concentrations result in exaggerated relative differences with respect to the base case.

Coarse Particulate Matter Concentrations

Examination of Figures 3.2-3.3 provides insight into the sensitivity of coarse particulate matter concentrations to changes in the wet and dry deposition of coarse particulate matter. First, one should note the location of two maxima that appear in base case concentration field (depicted by the diamond transect) of the absolute graphs. The first is located in grid cell 3 over northern Georgia and has a concentration of nearly 1.4 ug/m^3. The second maximum,which is located in grid cell 9 over western Pennsylvania, is the largest and has a concentration of 1.9 ug/m^3. A sharp gradient in the concentration field occurs after this maxima as values fall off quickly to less than 1.0 ug/m^3 as the transect enters Canada. Secondly, examination of the figures also reveals that increasing either the wet or dry deposition of the coarse particulate matter results, as expected, in a decrease in the concentration, and that this decrease is more pronounced in the case of wet deposition. Likewise,

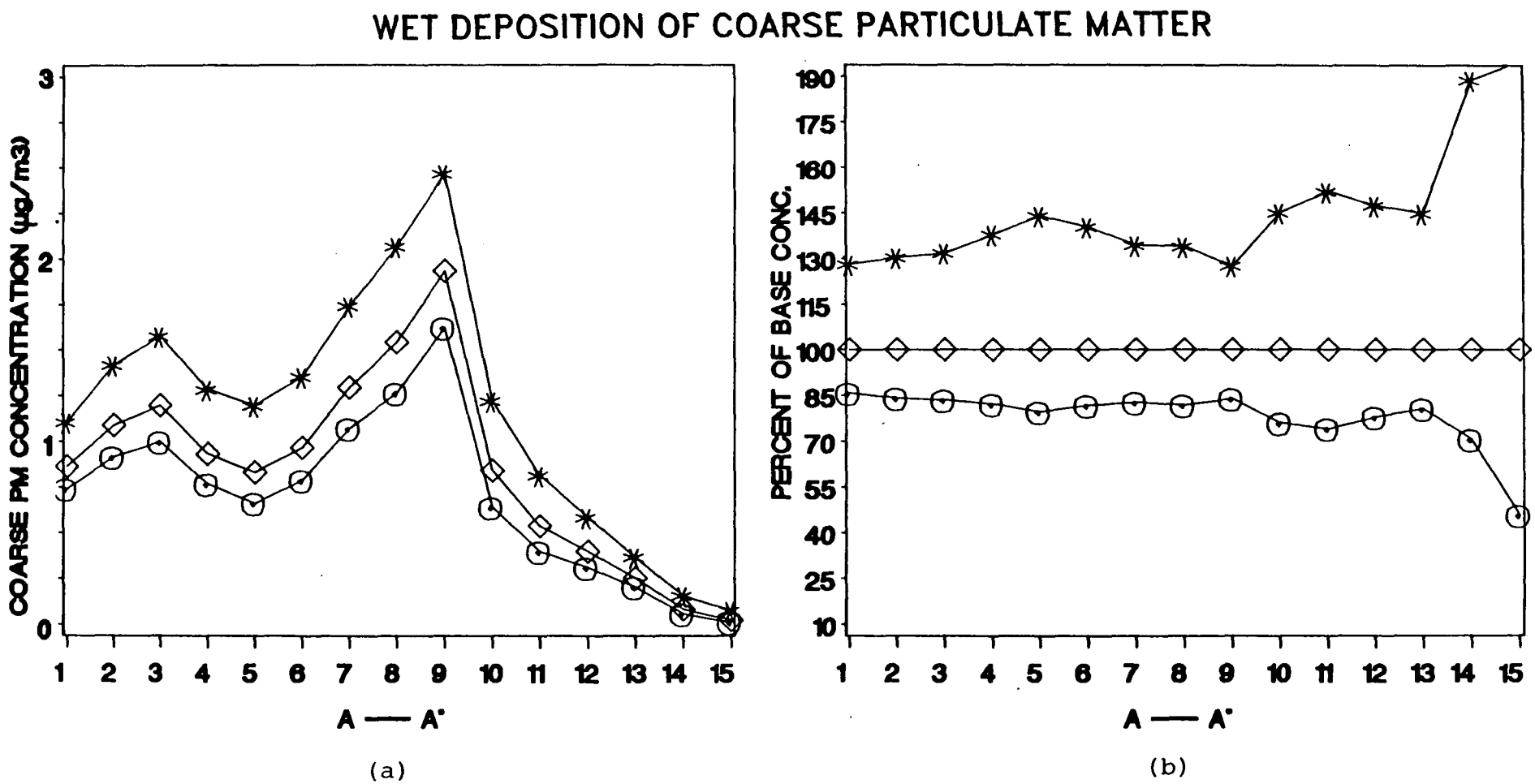


Figure 3.2 Absolute (a) and relative (b) sensitivity of coarse particulate matter concentration to changes in the wet deposition rate of coarse particulate matter. (Asterisk - 50% normal wet deposition, Diamond - 100% normal wet deposition, Circle - 150% normal wet deposition.)

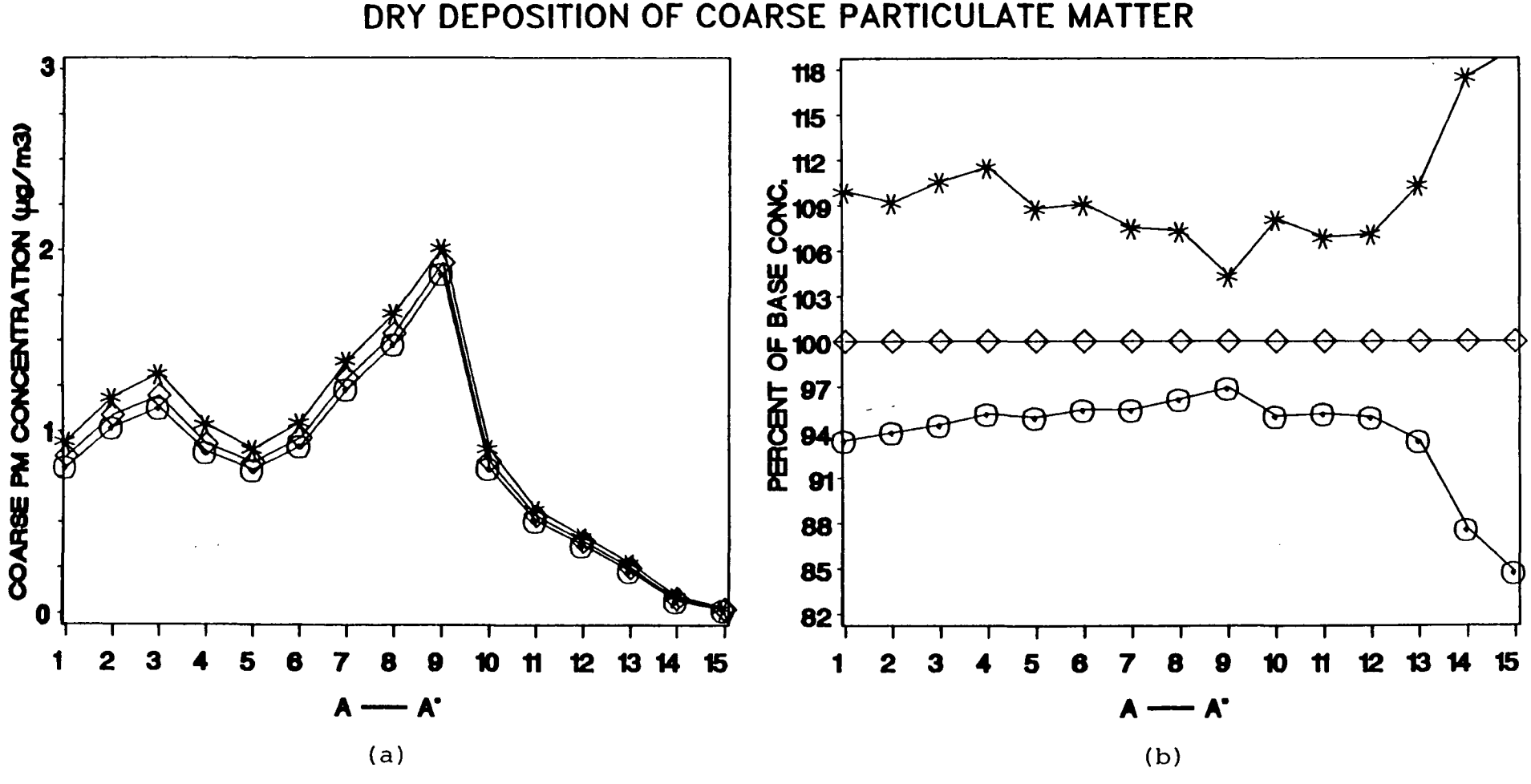


Figure 3.3 Absolute (a) and relative (b) sensitivity of coarse particulate matter concentration to changes in the dry deposition rate of coarse particulate matter.

decreasing either the wet or dry deposition of coarse particulate matter results in increased concentrations.

It is interesting to note that the changes in the magnitudes of the concentration patterns proved to be non-linear. That is, the changes in the model output are not directly proportional to changes in the input, and in most cases are far less than 50%. This non-linearity can in part, be attributed to the ability of the parameters to compensate for a given increase or decrease in a specific parameter. This compensation will to some degree reduce the response of the model to the forced variation.

This non-linearity is also evident in that the difference between the base case and the low deposition rates (both wet and dry) is considerably larger than that between the base case and the high deposition rates. This is due to the different "efficiencies" exhibited by each of the parameterizations. Each parameter has a maximum rate, which once exceeded, will produce no further changes in the model's simulations. This threshold value is more easily attained for the more efficient of the parameterizations, which include all of the wet deposition rates, the dry deposition of coarse particulate matter, and to a lesser degree the transformation rate of SO_2 into $SO_4^=$. Therefore, increasing these efficient parameters by 50% will often result in this threshold rate being exceeded, thereby limiting the impact on the concentration.

This phenomenon is well illustrated in the graphs showing the relative changes in the concentration field. Examination of the wet deposition graph Fig 3.2.b, shows that for a 50% decrease in the wet deposition, the concentration increases an average of

30 to 50%, but that for a 50% increase in the wet deposition, the concentration only decreases an average of 15 to 25%. Similar trends are evident, but to a lesser degree, with the dry deposition graph as seen in Fig 3.3.b. For a 50% decrease in dry deposition, the concentration increases an average of 5 to 10%, but for a 50% increase in dry deposition, the concentration only decreases between 3 and 6%.

Another interesting feature of the graphs, which is evident throughout all of the analysis, is that the basic spatial pattern of the concentration appears to remain the same. That is, the location of the relative maxima and minima remain the same and do not shift up or down the transect.

Fine Particulate Matter Concentrations

Examination of Figures 3.4-3.8, which depict the sensitivity of fine particulate matter concentration to changes in the wet and dry deposition of SO_2 and fine particulate matter, as well as to changes in the transformation rate of SO_2 into $SO_4^=$, reveals many of the same characteristics as noted with the coarse particulate matter. First of all, two local maxima are again evident in the concentration field. The first is located in the second grid cell which falls over northwestern Alabama and has a value of 3.0 ug/m^3. The second and largest maxima, which has a value of 4.3 ug/m^3, is located in grid cell number nine, which is over western Pennsylvania. As was seen in the concentration field of the coarse particulate matter, the concentration of fine

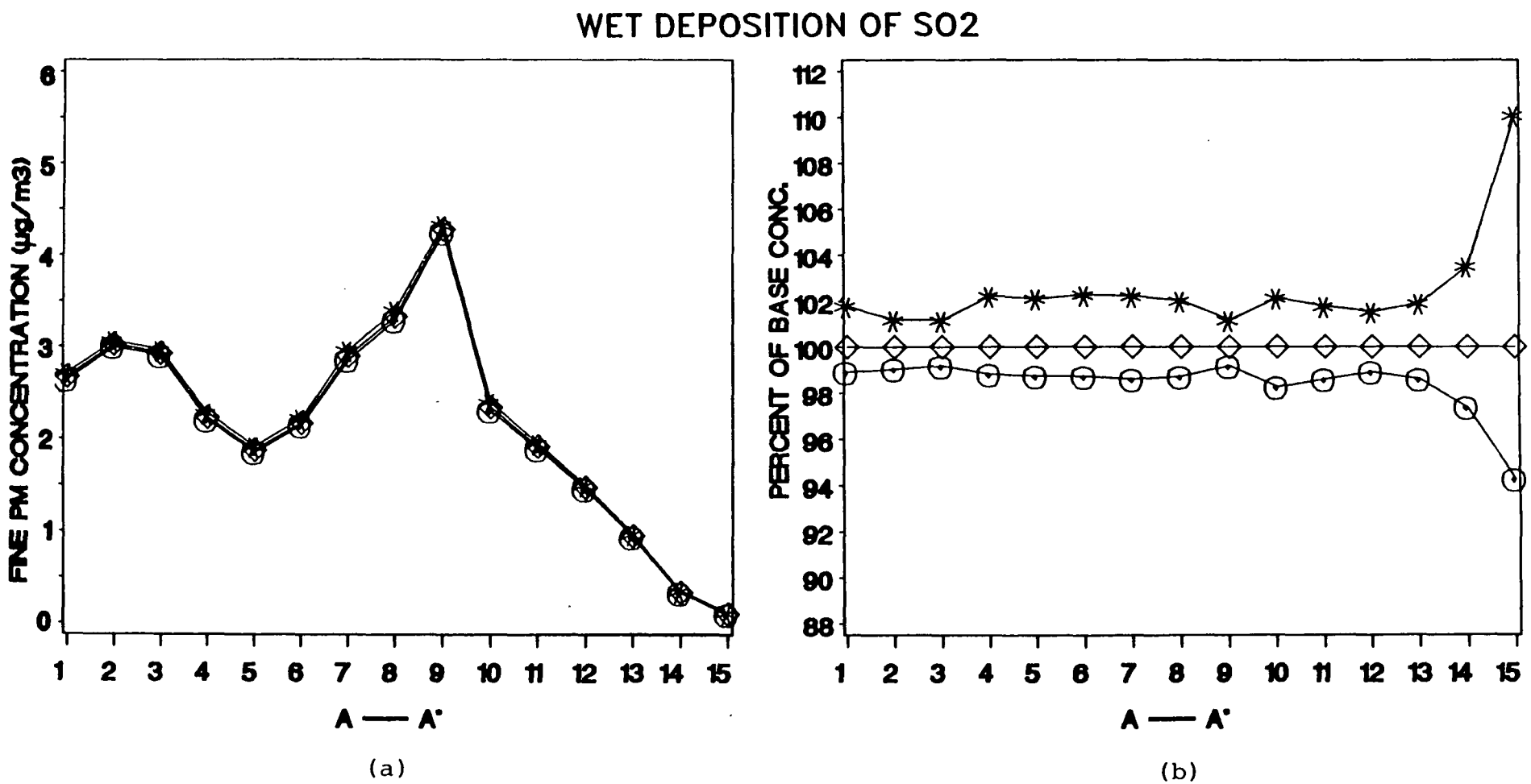


Figure 3.4 Absolute (a) and relative (b) sensitivity of fine particulate matter concentration to changes in the wet deposition rate of SO_2.

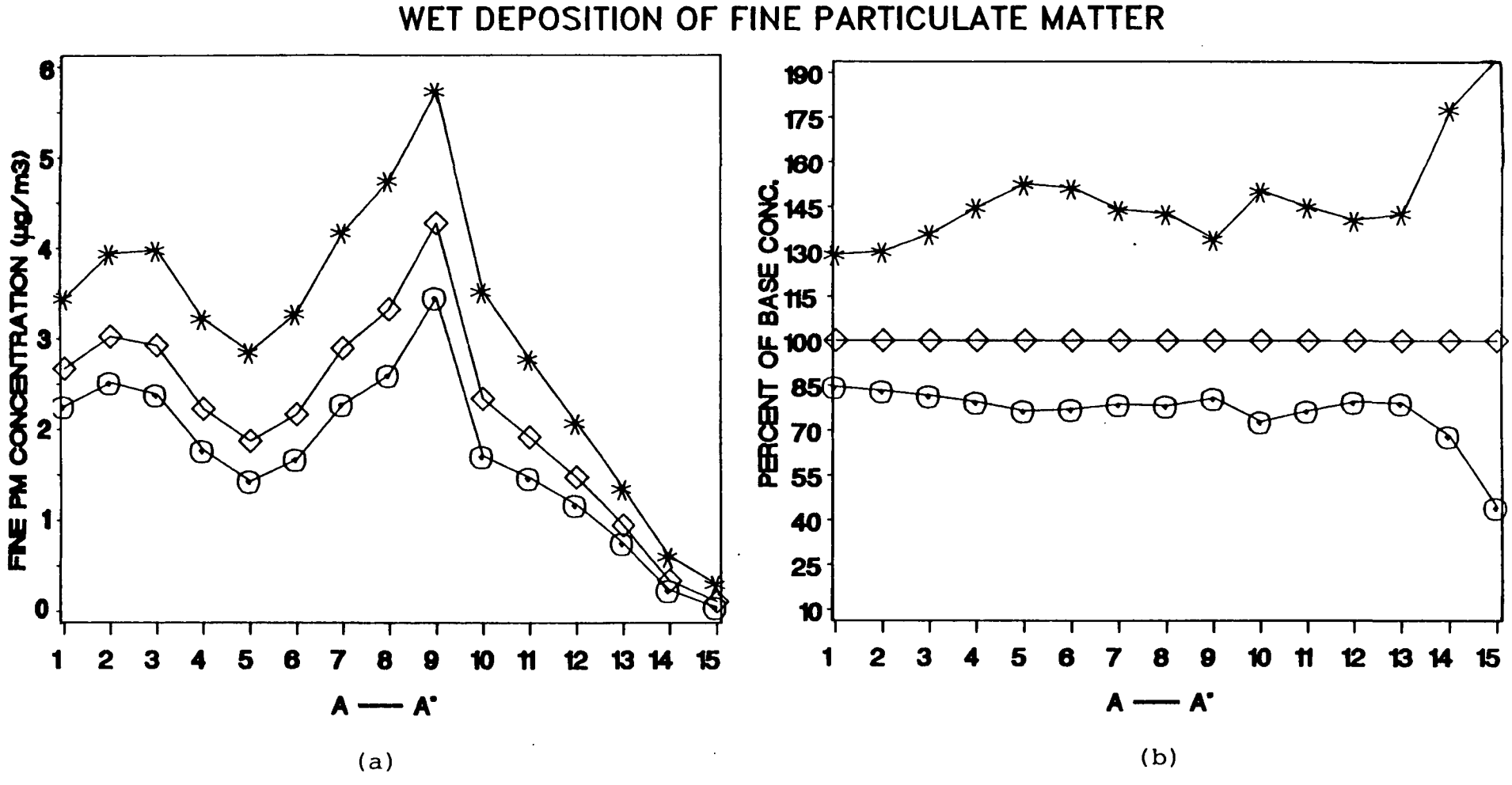


Figure 3.5 Absolute (a) and relative (b) sensitivity of fine particulate matter concentration to changes in the wet deposition rate of fine particulate matter.

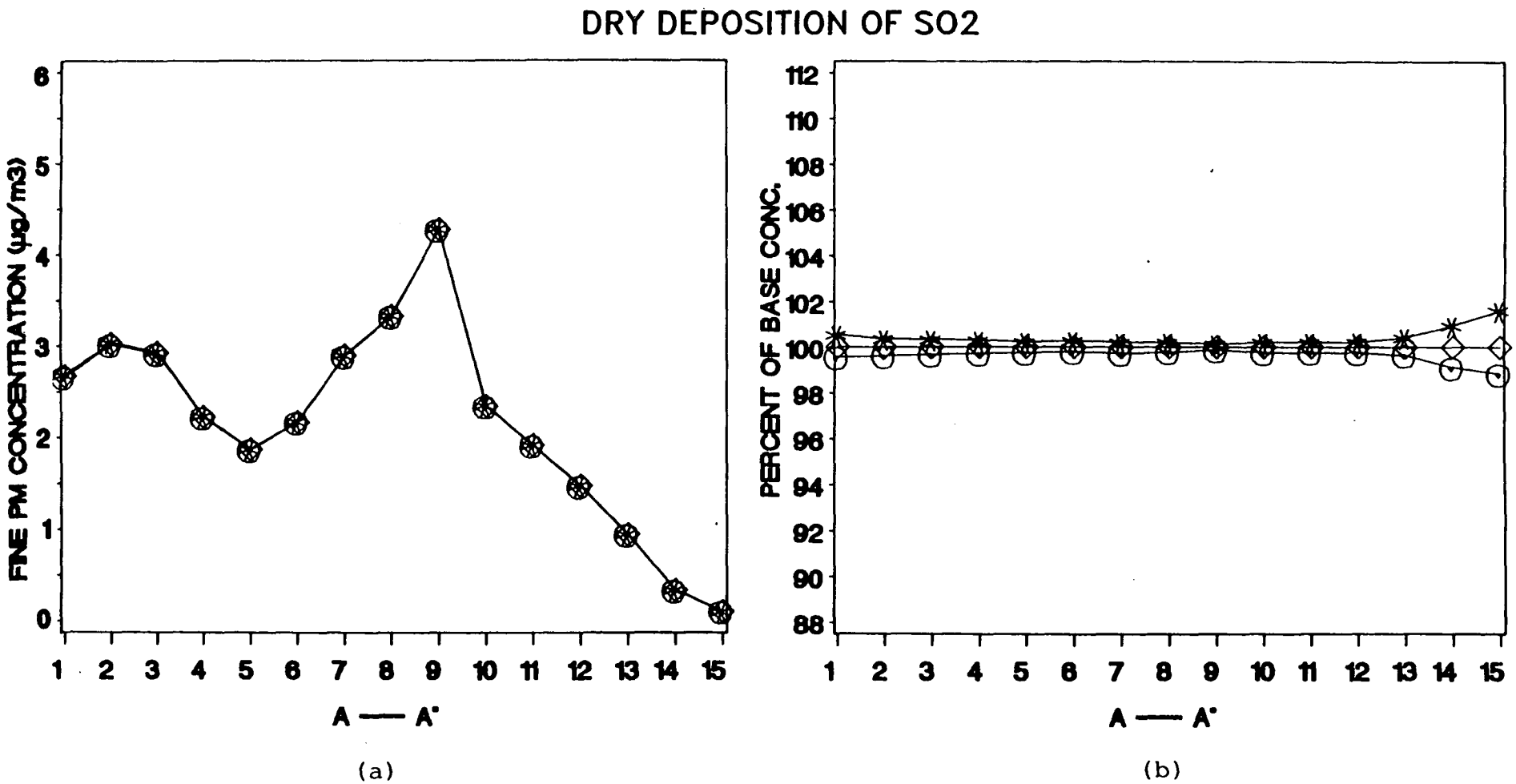


Figure 3.6 Absolute (a) and relative (b) sensitivity of fine particulate matter concentration to changes in the dry deposition rate of SO_2.

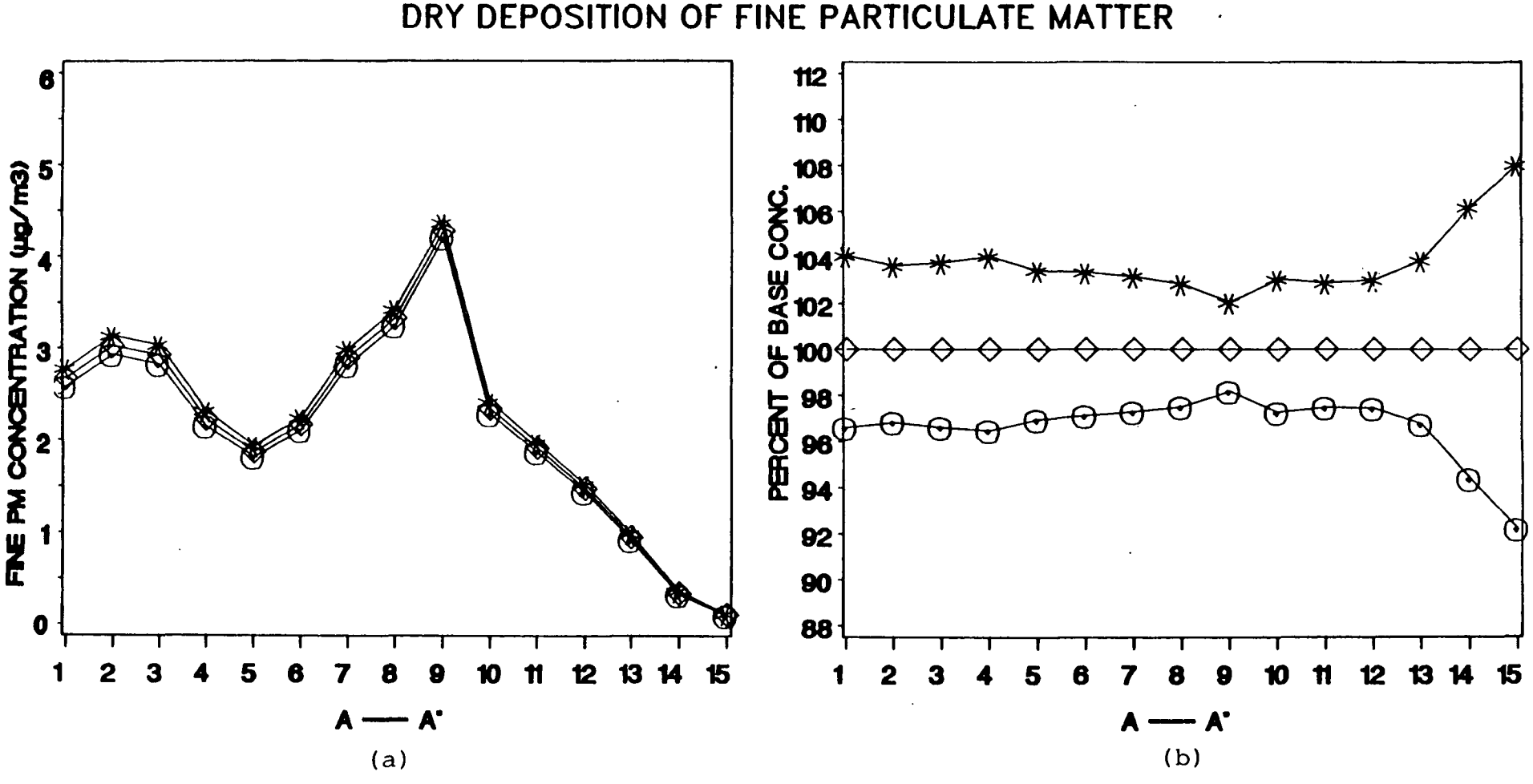


Figure 3.7 Absolute (a) and relative (b) sensitivity of fine particulate matter concentration to changes in the dry deposition rate of fine particulate matter.

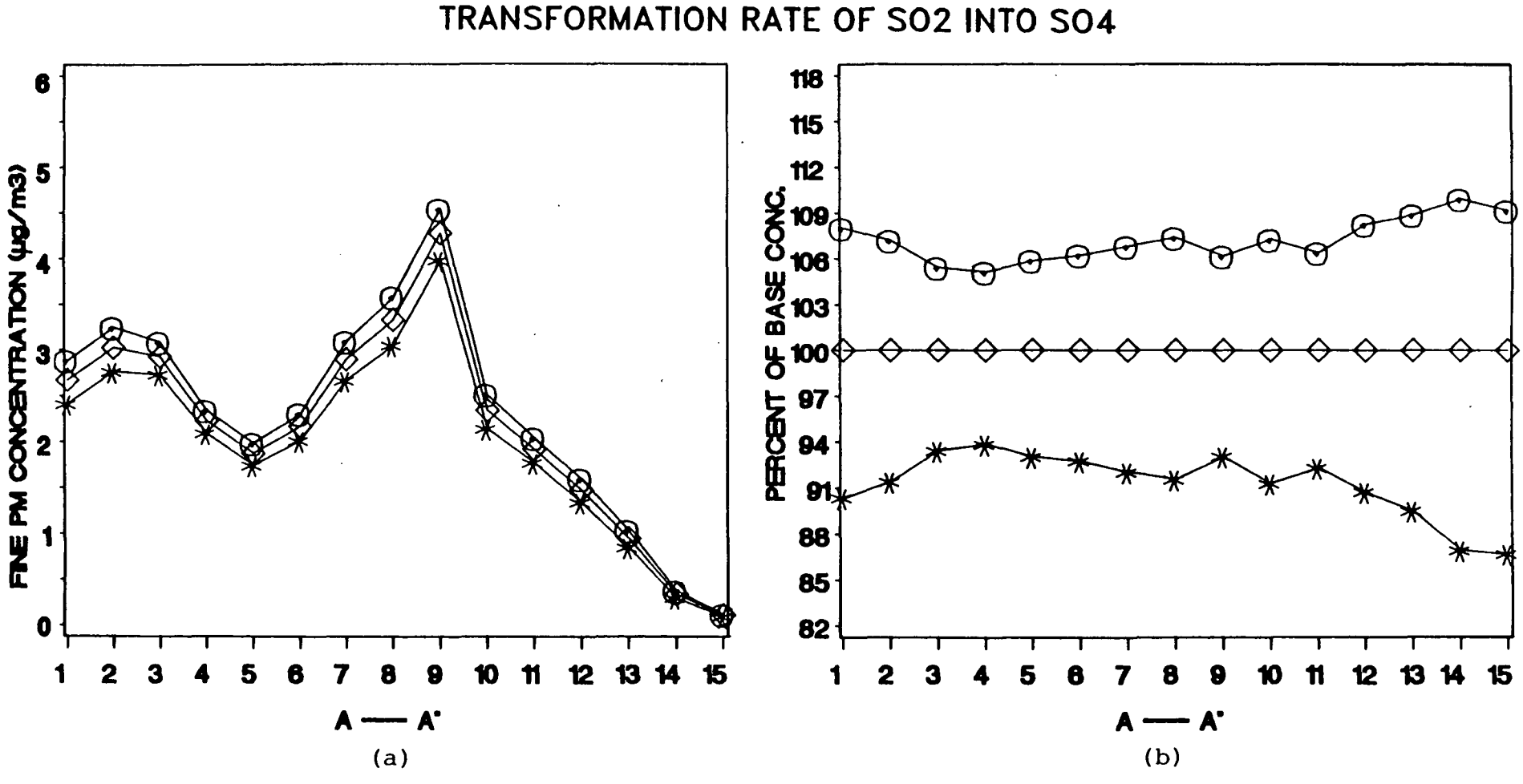


Figure 3.8 Absolute (a) and relative (b) sensitivity of fine particulate matter concentration to changes in the transformation rate of SO_2 into $SO_4^=$.

particulate matter falls off rapidly as the transect enters southeastern Canada.

The sensitivity of fine particulate matter concentration to wet deposition of SO_2 (which is a precursor to $SO_4^{=}$) and to fine particulate matter each exhibit non-linear behavior. The influence of SO_2 wet deposition, however, proves to be minimal as seen in Figures 3.4 a and b. Examination of the relative graph shows, that with the exception of the last two grid cells, at most a 3% change in the concentration field occurs given a 50% change in the SO_2 wet deposition field.

As expected, the wet deposition of fine particulate matter had a much larger impact upon the concentration field as seen in Figures 3.5 a and b. For a given 50% increase in the wet deposition of fine particulate matter, the concentration decreased an average of 15 to 30%, whereas a 50% decrease in the wet deposition resulted in a 30 to 50% increase in the concentration.

Examination of the dry deposition graphs for both SO_2 and fine particulate matter, as seen in Figures 3.6 - 3.7, reveals that the sensitivity of the fine particulate concentration to these less "efficient" parameterizations proved to be linear. That is, the difference found between the base case and the low dry deposition rates is equivalent to the difference found between the base case and the high deposition rates. This linearity is evidenced through the "mirror image" effect seen in the relative graphs about the base case line. The influence of SO_2 dry deposition on the concentration of fine particulate

matter proved to be all but non-existent as seen in Figure 3.6. A +/- 50% change in the SO_2 dry deposition resulted in at most a +/- 1% change in the concentration field. The impact of fine particulate dry deposition on the fine concentration field, though small, is more noticeable as seen in Figure 3.7. Inducing a +/-50% change in the dry deposition of fine particulate matter resulted in a 3 to 6% change in the concentration field.

And finally, as seen in Figures 3.8, the sensitivity of fine particulate matter concentration to changes in the transformation rate is both non-linear and rather significant. A 50% increase in the transformation rate increases the concentration by an average of 5 to 10%, while a 50% reduction in the transformation rate results in a 6 to 12% decrease in the concentration.

The results of separately introducing +/- 50% changes into three major parameterizations (wet deposition, dry deposition, and transformation rate) involving the simulation of fine and coarse particulate matter concentrations has been examined. Although the results are preliminary, several important conclusions can be drawn:

- Simulated concentrations of fine and coarse particulate matter are by far most sensitive to variations in the wet deposition rates of fine and coarse particulate matter, respectively. (For a given +/- 50% change in the wet deposition, a +/- 15 to 45% change occurs in the concentration). However, concentrations of fine particulate matter proved to be quite insensitive to wet deposition of SO_2 (+/- 1 to 4%).

- Concentrations of coarse particulate matter are somewhat sensitive to dry deposition of coarse particulate matter (+/- 5 to 10%). Concentrations of fine particulate matter are, however, less sensitive to dry deposition of fine particulate matter (+/- 2 to 6%), and are in fact highly insensitive to dry deposition of SO_2 (+/- 1%).

- Concentrations of fine particulate matter proved to be somewhat sensitive to the transformation rate of the precursor SO_2 into $SO_4^=$ (+/- 5 to 10%).

SECTION 4

PRELIMINARY MODEL PERFORMANCE EVALUATION

In order to perform an adequate model performance evaluation, three major components are necessary. First, a complete and detailed meteorological input data set that accurately simulates the atmospheric process that are pertinent to the model simulations is necessary. Second, a comprehensive emissions input data set which emulates both the anthropogenic as well as the natural emissions found in the model's domain is needed. The third, and perhaps most important component, is a complete evaluation data set that can be used to validate each of the output parameters simulated by the model over compatible spatial and temporal scales. Unfortunately, for reasons that will be discussed later in this section, only the input meteorological data can be deemed adequate at this time. Inadequacies inherent to both the emissions input data set and the model evaluation data set limit the scope of this evaluation, therefore it must be considered preliminary at this time.

RELMAP was run for the three month period of July, August and September, 1980 in order to simulate a summer season using meteorological data obtained from the National Climatic Data Center located in Asheville, N. C. Included in the meteorological data are gridded 12-hourly surface and 850 mb wind data and

hourly precipitation data. Gridded input emissions data were obtained from the National Acid Precipitation Assessment Program (NAPAP) Version 5.0 Emission Inventory. Simulated ambient air concentrations of fine and coarse particulate matter were then compared on a monthly and seasonal basis with monitoring data obtained from the Inhalable Particulate Network (IPN) data set. This section provides a brief overview of both the NAPAP Version 5.0 Emissions Inventory and the IPN data set and discusses the many inadequacies encountered when trying to incorporate them in this model evaluation.

Deficiencies in the NAPAP Version 5.0 Emissions Inventory

Version 5.0 of the 1980 NAPAP Emissions Inventory was selected for use in this evaluation because it represents by far the most comprehensive and highest quality emissions data set available. The Task Group on Emissions and Controls of the Interagency Task Force on Acid Rain was responsible for developing the inventory in order to support the modeling needs of NAPAP.

The emissions inventory contains point source emissions data for over 14,000 plants comprised of 52,000 source classification codes (SCC). Area source emissions are reported for 88 emission categories for over 3,000 counties in the contiguous U. S., and for 157 emission categories for the 10 Canadian provinces (Wagner et al., 1986.) In addition to the SO_2, $SO_4^=$ and fine and coarse particulate emissions of interest to this evaluation, data are

also available for NO_x, Pb, CO, HCl, HF, NH_3, VOC, and total hydrocarbons.

Unfortunately for this evaluation, the primary reason for developing the 1980 NAPAP emissions inventory was to provide an emissions data base for acid deposition research and modeling, not regional particulate modeling. Because of this, less emphasis was placed on the TSP inventory, resulting in numerous deficiencies in both the fine and coarse particulate emissions. The total annual emissions of TSP for the entire NAPAP grid area was estimated to be 74,192 ktons. Of this total, 42,617 ktons or 57.4% was emitted from U. S. sources, and 31,574 ktons or 42.6% was emitted from Canadian sources. Characteristics of the TSP emissions found within the boundary of the RELMAP grid and therefore used in this model evaluation are displayed in Figures 4.1 and 4.2.

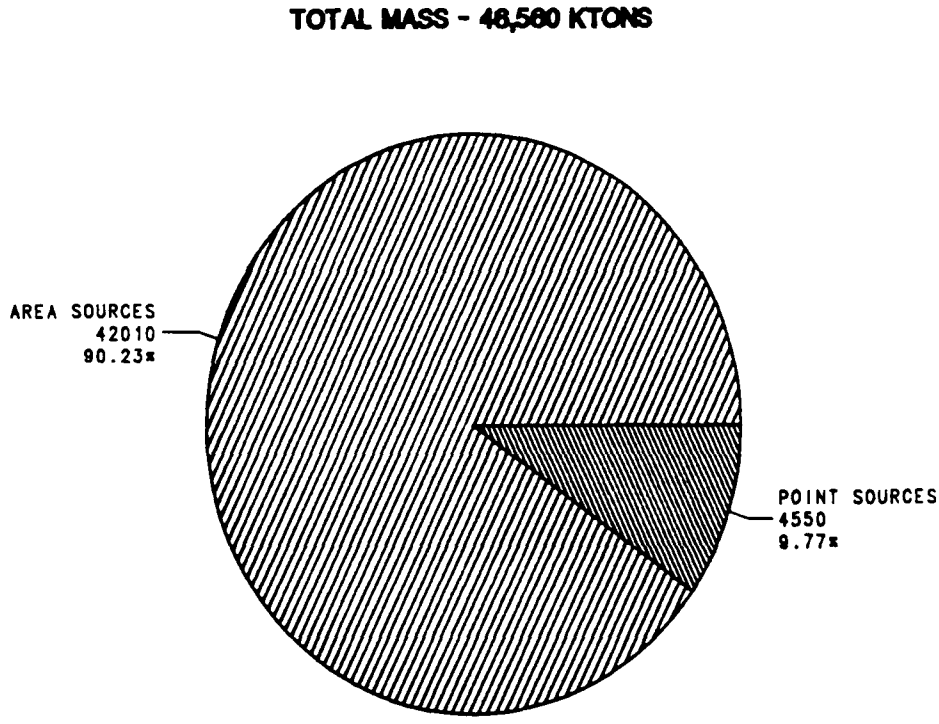
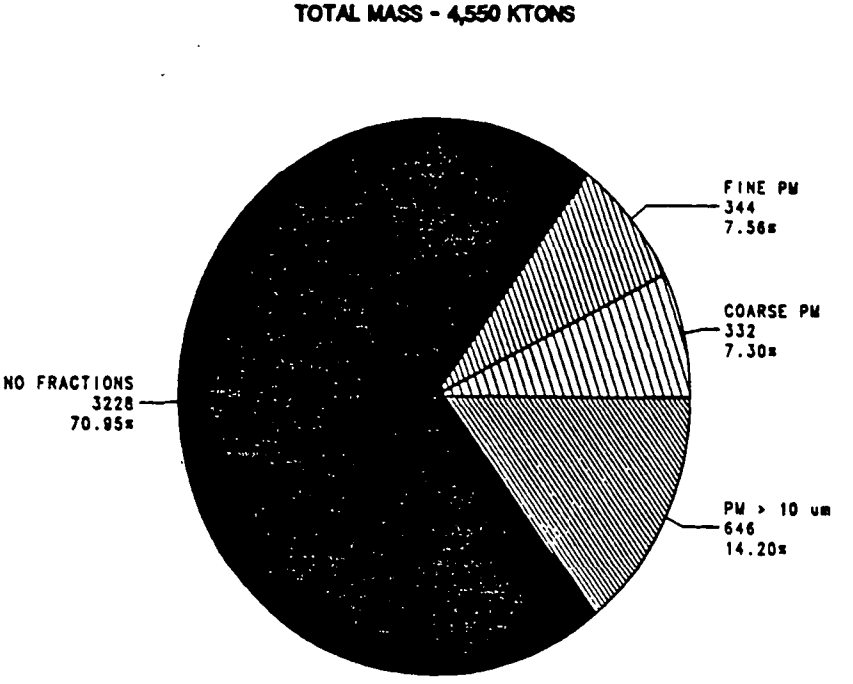


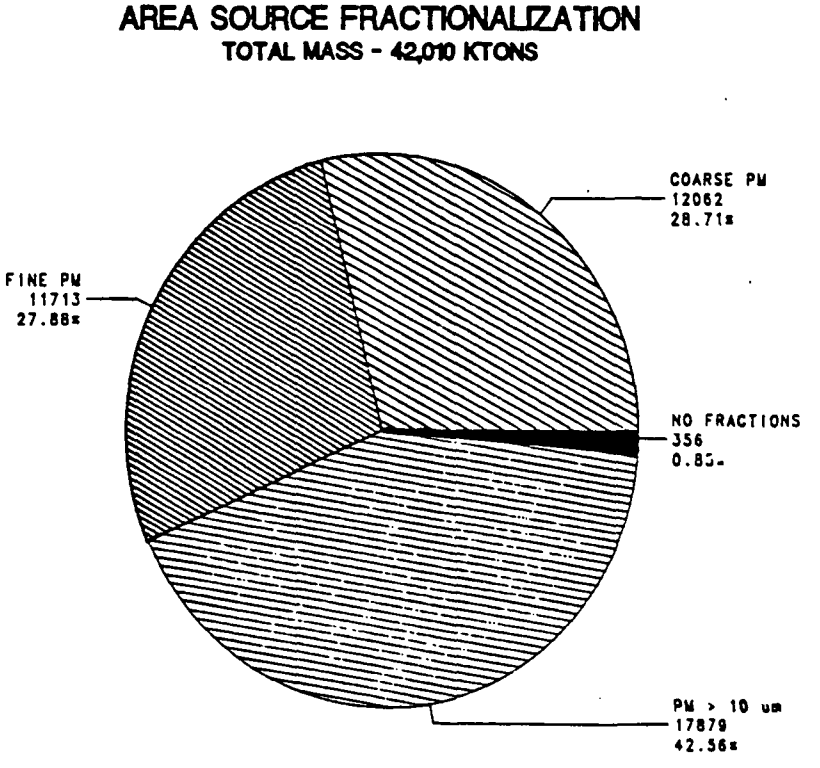
Figure 4.1 Area and point source emissions of TSP emitted within the RELMAP domain.

These figures illustrate the fractionalization of total TSP, as well as fine and coarse particulate matter. As seen in Figure 4.1, 46,560 ktons or 62.8% of the total NAPAP TSP inventory is emitted within the model's domain. Of this total, 90.23% can be attributed to area sources, and 9.77% can be attributed point sources. Figures 4.2 a and 4.2 b break these percentages down even further. Of the 4,550 ktons of TSP attributed to point sources, 14.20% are emitted as particles with diameters larger than 10 um, 7.56% are emitted as fine particles, 7.29% are emitted as coarse particles, and 70.95% cannot be fractionalized. This last percentage illustrates one of the two major deficiencies of the NAPAP TSP inventory. A large percentage of the many point source categories designated by NAPAP do not have particle size distributions. Because of this, more than 3 million tons of the TSP emitted from point sources can not be fractionalized, or broken down into the respective size categories.

Examination of the area source fractionalization reveals that of the 42,010 ktons of TSP attributed to area sources, 28.71% are emitted as coarse particles, 27.88% are emitted as fine particles, 42.56% are emitted as particles with diameters larger than 10 um, and 0.85% cannot be fractionalized. In a situation similar to that seen with the point sources, fractionalization was only possible with 64 of the area source categories designated by NAPAP. Over 356 ktons of TSP from the area sources were omitted because 24 of the U. S. and 93 of the Canadian categories could not be fractionalized. When combined with the point source emissions, a total of 3,584 ktons or 7.70%



(a.)



(b.)

Figure 4.2 Point source (a.) and area source (b.) fractionalization of TSP emitted within the RELMAP domain.

of the TSP emissions found in the NAPAP inventory cannot be fractionalized. OAQPS is currently updating their SCC inventory, but until this is completed, these non-fractionalized emissions cannot be used as input into the model, which will have detrimental effects on the model's performance.

Another, even more serious deficiency found with the NAPAP inventory is the omission of many of the "open" sources of TSP. Open source emissions, which are defined as sources of air pollution too great in extent to be controlled by enclosure, are extremely difficult to estimate. Open sources of TSP include paved and unpaved roads, agricultural tilling, wind erosion, construction activity, forest fires (wild and prescribed) and mining operations. Unfortunately, only roads (paved and unpaved) and forest fires are included in the inventory. The omission of agricultural tilling, wind erosion, construction and mining sources from the inventory reflected the different methodologies employed by Canada and the U. S. Originally, the Canadian inventory of TSP (which included 157 area source emissions) included all of the open sources omitted above; however, because there were no counterparts in the U. S. for agricultural tilling, wind erosion, construction and mining, these major sources were dropped from the inventory altogether in an effort to be consistent.

As an illustration of the magnitude of this problem, estimates of the amount of these omitted open sources for states within the RELMAP domain were calculated and are presented in Table 4.1. Unfortunately, these totals, which were derived from Evans and Cooper (1980), are based upon 1976 data and exclude

Canadian provinces. The final, annual estimate of 255,646 ktons, which again excludes the Canadian provinces within the model's domain, is more than five times larger than the total annual TSP emissions accounted for in the NAPAP inventory!

Table 4.1 Estimates of Omitted Open Source Emissions of TSP for States Within the RELMAP Domain.

SOURCE	TSP EMISSIONS (KTons)
Agricultural Tilling	31,446
Wind Erosion	206,776
Construction	15,137
Mining	2,287
TOTAL	255,646

It should be noted, however, that open sources tend to emit larger particles than most anthropogenic sources, and that open sources tend to be located in remote areas, far removed from population centers and TSP monitoring sites. Because of this, Evans and Cooper estimated that a ton of open source emissions has between $1/10^{th}$ and $1/40^{th}$ the impact as does one ton of anthropogenic sources at a TSP monitoring site. Even if this reduction is applied, between 6,400 and 25,560 ktons, of open sources that are emitted within the states are being omitted from the NAPAP inventory. Comparison with the NAPAP inventory estimates of TSP for the U.S. alone, supports the claim that open

sources contributions are equivalent under the most conservative of estimates, to the anthropogenic sources, yet most are not included in the inventory.

A third deficiency, which is as detrimental to the evaluation as the second, is the wide discrepancy observed between the estimates of open source emissions due to unpaved roads. Over 70% of the total TSP emissions in the NAPAP inventory is attributed to unpaved roads. Unfortunately, the total estimated by NAPAP for this category is much lower than other independent estimates. For instance, Evans and Cooper estimate that in the U.S. alone, over 170,000 ktons of TSP emissions are emitted from unpaved roads, which is almost an order of magnitude higher than the NAPAP total.

Because of the number and seriousness of these deficiencies, any model performance evaluation using the NAPAP inventory as a source ofTSP emissions must be considered preliminary at best. Until emissions of TSP are given the same consideration as those of SO_2, $SO_4^=$ and other detrimental pollutants, modeling of fine and coarse particulate matter will continue to lag behind the other modeling efforts being undertaken today.

Deficiencies In The Inhalable Particulate Network Data Set

The Inhalable Particulate Network (IPN) was developed and implemented by the Environmental Monitoring Systems Laboratory (EMSL) in conjunction with the Office of Air Quality Planning and Standards (OAQPS). The IPN was designed to collect size-specific

particulate data in anticipation of the 1977 Clean Air Requirement Act, which called for a reappraisal of the National Ambient Air Quality Standard for particulate matter. One reason for this reappraisal was a shift in emphasis from Total Suspended Particulate matter (TSP), which ranged in size from 0.0 to 50.0 um, to the smaller Inhalable Particulate (IP), which ranges in size from 0.0 to 15.0 um.

The IPN became operational during April 1979, when 57 sites located throughout the United States went on-line using hi-vol, dichotomous and size selective inlet samplers to collect data on TSP, FINE-10 (0-2.5um), and COARSE-15 (2.5-15um) particulate matter. The network eventually grew to 157 sites, when in 1981, EPA's OAQPS recommended that the revised primary standard for ambient air concentration should be based on a 10 um criteria rather than the 15 um. The 15 um limit, which had been suggested by Miller et al. (1979), had been the subject of debate since the network's conception. EPA's decision, which was based upon the recommendations of the Clean Air Science Advisory Committee and the International Standards Organization Task Group, initiated efforts toward the deployment of 10 um size specific samplers at 39 sites in the IPN during 1982. This effort, however, was too late to provide the necessary data for this model evaluation.

Unfortunately, of the 157 IPN sites that were in operation at one time or another, only 14 were spatially and temporally compatible with this evaluation. Table 4.2 provides a list of these sites along with their respective IPN site numbers, locations and land use categories. Figure 4.3 illustrates the location of these sites.

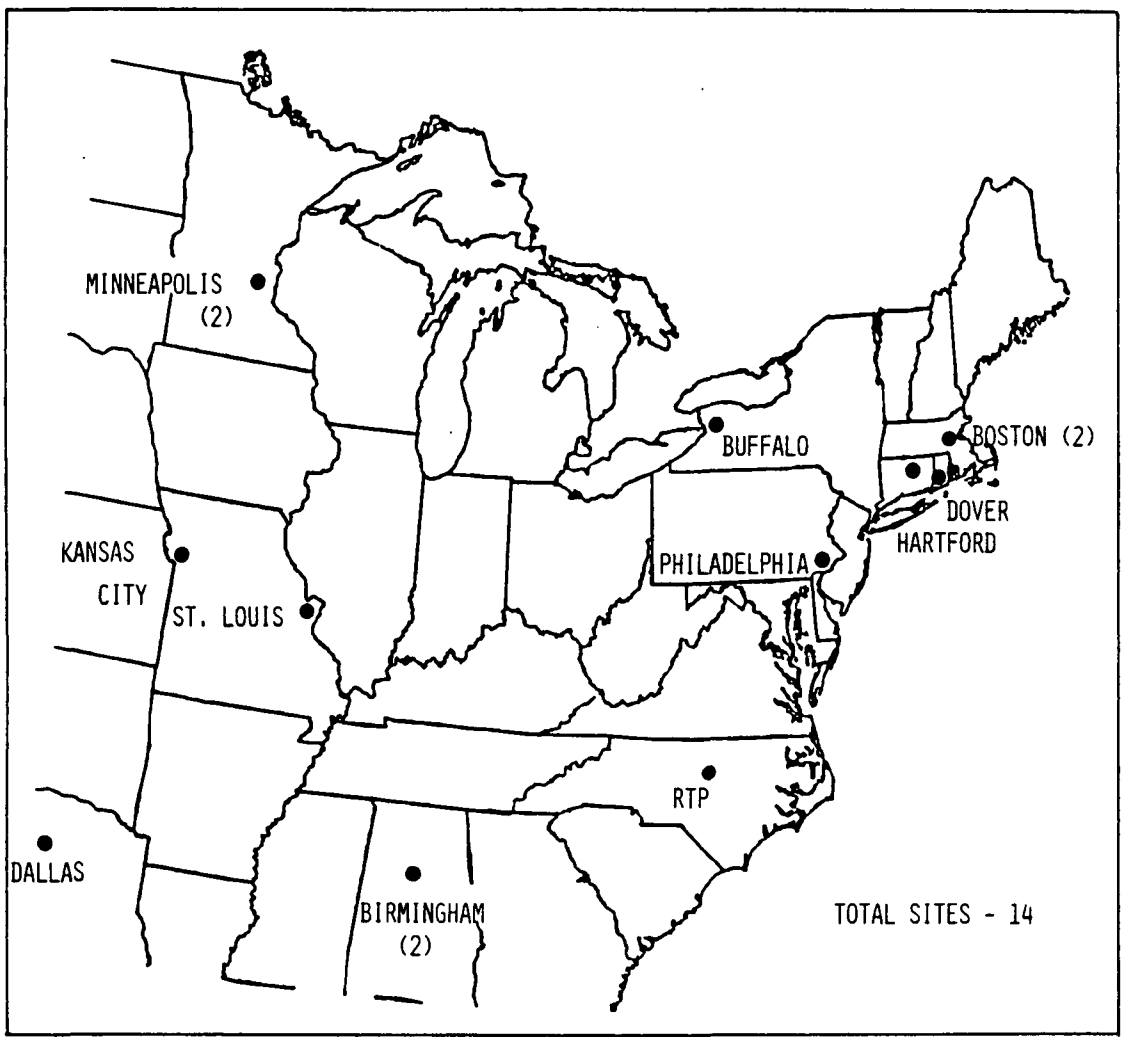


Figure 4.3 Inhalable Particulate Network sites used in the preliminary model evaluation.

Table 4.2 Inhalable Particulate Network sites used in the preliminary model evaluation.

NAME	SITE #	STATE	LOCATION	LAND-USE
Huffman	010570001A07	AL	Suburban	Residential
Mtn. Brook	012540001A07	AL	Suburban	Residential
Hartford	070420003A07	CT	Center City	Commercial
Dover	080020001A07	DL	Center City	Commercial
Boston (Fire Hq)	220240012A07	MA	Suburban	Commercial
Boston (S Cen)	220240013A07	MA	Center City	Commercial
Minneapolis (HS)	242260049A07	MN	Center City	Residential
Minneapolis (Nic)	242260051A07	MN	Center City	Commercial
St. Louis	260030001A07	MO	Suburban	Commercial
Kansas City	262380002A07	MO	Center City	Commercial
Buffalo	330660003A07	NY	Center City	Residential
RTP	341160101A07	NC	Rural	Commercial
Philadelphia	397140024A07	PA	Suburban	Commercial
Dallas	451310050A07	TX	Center City	Commercial

A total of 41 sites were located outside the model's domain, and 62 sites did not come 'online' until after the evaluation period. Of the 54 remaining sites, 33 had insufficient data (i.e. less than 10 observations during the three month evaluation period), and 7 were located in areas that were classified as industrial. Unfortunately, of the 14 sites that were spatially and temporally compatible, 6 were co-located sites, (i. e. they were located within the same city and grid cell) which further reduced the spatial representiveness of the evaluation data set. These deficiencies, as well as others, are elaborated upon below.

In order to adequately evaluate a regional scale model such as RELMAP, which has a 1^{o} by 1^{o} grid cell resolution, one would ideally have a monitoring network made up of remote locations that have the same spatial and temporal resolution as the model. Unfortunately, the IPN was designed primarily to characterize urban scale concentrations of suspended particulate matter, since the attainment of air quality standards is evaluated over this scale (Watson et al.,1981). Because of this, an overwhelming majority of the IPN sites are classified as either center city or suburban, where the dominant land use is described as either industrial, commercial or residential. In fact, of the 157 sites that make up the network, only 5 are classified as remote, and 9 classified as rural. Of these 14 sites sites available for the evaluation, only one, the Research Triangle Park (RTP), NC is classified as rural.

In a study performed in the Detroit area, Wolff et al., (1984) concluded that <u>regionally</u> emitted emissions generally

dominate the ambient concentration of fine particulate matter over local emissions. But, they also concluded that ambient concentrations of coarse particulate matter were dominated by local sources at all of the sites. With this in mind, a further criteria was established for sites used in this evaluation in that they must not be industrial in nature, which further reduced the number of available evaluation sites.

With few exceptions, the hi-vol, dichotomous and SSI samplers used in the IPN were only activated once every six days, at which time 24-hour average ambient air concentrations were recorded from midnight to midnight (LST). These sixth-day observations resulted in a dearth of data, which in turn made the model evaluation very difficult and preliminary at best. The maximum number of 24 hour observations available for the three month evaluation period was 16, with 6 of these observations occurring in July and September, and 5 in August.

This limited number of observations was further depleted when the amount of "down time" for each site was considered. Of the 14 stations used in the evaluation, only two, Philadelphia and Mtn. Brook, Alabama, had the full allotment of 16 observations. The 12 remaining sites averaged between 12 and 14 with a minimum of 10. Using such a temporally inconsistent data set makes the observations very susceptible to extremes caused by local sources. The tremendous variability exhibited by the observed data, whether real or artificial, cannot be modeled by a regional-scale, long term (monthly) model such as RELMAP. This incompatibility is best illustrated by Figures 4.4 and 4.5, which presents the observed and simulated fine and coarse

particulate matter concentrations for Hartford, Connecticut during the three month evaluation period. On the abscissa, one finds the date, which ranges from July 1, to October 1, 1980, while the ambient air concentrations are on the ordinate. At best, the observed data is inconsistent, with four of the 16 observations missing. Another unfortunate characteristic of the observed data is its tremendous variance. Fine particulate concentrations range from 6 to 45 ug/m^3, while the coarse concentrations range from 4 to 17 ug/m^3. Such variance, which may be an indication of local sources, is impossible to simulate by the model.

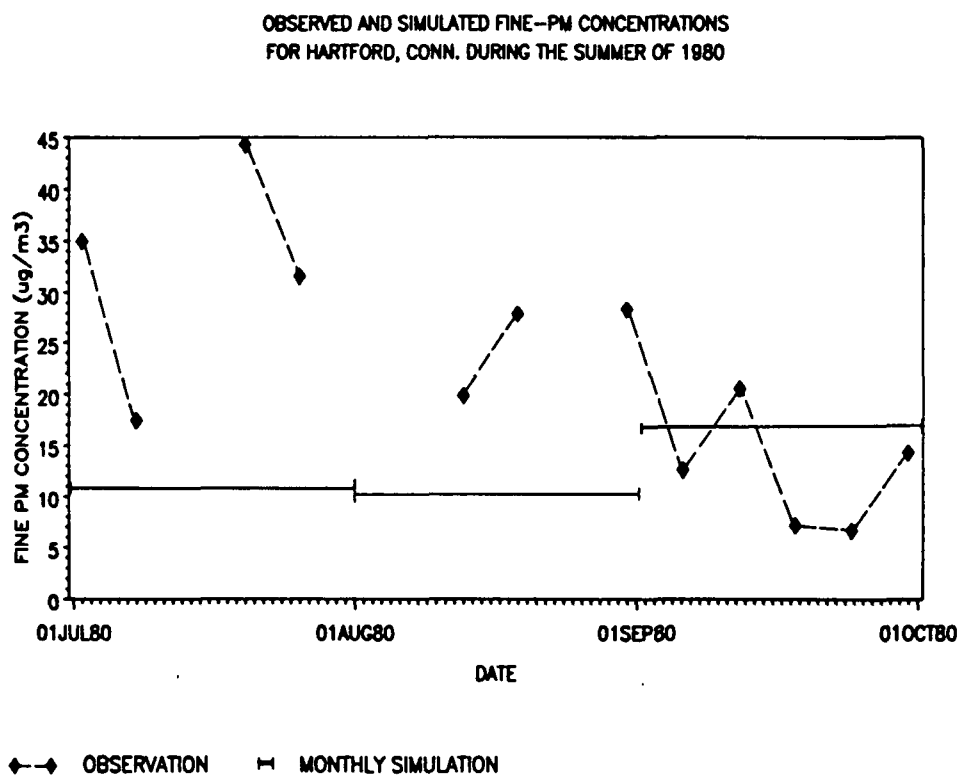


Figure 4.4 Temporal depiction of the observed and simulated fine particulate matter concentrations for Hartford, Conn., for the summer of 1980.

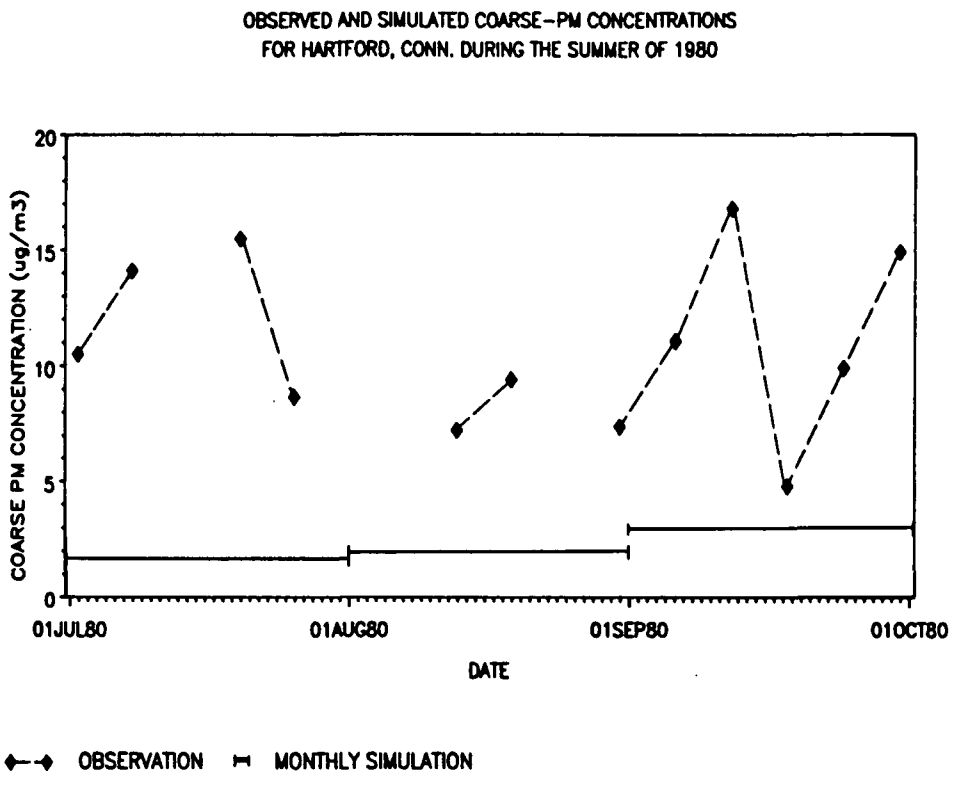


Figure 4.5 Temporal depiction of the observed and simulated coarse particulate matter concentrations for Hartford, Conn., for the summer of 1980.

As discussed in the previous section, when the emphasis was shifted from COARSE-15 to COARSE-10 in 1982, dichotomous samplers were incorporated into the IPN which measured COARSE-10. Unfortunately, this transition occurred too late to provide direct measurement of COARSE-10 needed for this evaluation. However, several methods have recently been developed that have allowed full utilization of the COARSE-15 data as a substitute for the COARSE-10 data. Rodes et al. (1984) examined the relationships between PM-15 (particles less than or equal to 15 um) and PM-10 (particles less than or equal to 10 um) data at eight cities located throughout the United States, and found that

PM-10 and PM-15 concentrations exhibited a very strong linear relationship, making it possible to predict one from the other. Correlations between the measurements at the eight sites, which included industrial, rural as well as suburban locations, ranged between 0.93 and 0.98. The ratio of PM-10/PM-15 was also consistent, ranging from 0.75 to 0.96, and averaging 0.85. It should be noted that the lowest value (0.75) was recorded at the only western site (Phoenix), and that if this outlier is removed from the data set, an average ratio of 0.87 would result.

In a similar study, Pedco Environmental, Inc. et al. (1984), examined, tested and evaluated 13 different methods in which PM-10 and PM-15 could be estimated from PM-15 and TSP, respectively. Among their final recommendations for estimating PM-10 from PM-15 data was to use a PM-10/PM-15 ratio of 0.88 in the eastern states and 0.77 in the western states. This method, which produced the smallest standard errors of any tested by Pedco Environmental, Inc. et al., (1984), and which was in very good agreement with Rodes et al.,(1984), was selected for use in this evaluation. Subsequently, all of the PM-15 data were converted into PM-10 data using the 0.88 ratio. The Coarse-10 fractions were then determined by subtracting the Fine-10 (which is the same as the Fine-15) fraction from the PM-10 total.

Model Evaluation for the Summer of 1980

RELMAP was run on a monthly basis for July, August and September, 1980 in order to produce monthly and seasonal

simulations of concentrations and wet and dry depositions of fine and coarse particulate matter. The monthly and seasonal simulated values of fine and coarse concentrations (expressed in ug/m^3) were then compared to the 14 compatible sites from the IPN. The number of stations used each month in the evaluation varied depending upon the number of which met the minimum observation requirement of 3 observations/month, or 10 observation/summer season. July and September had 13 stations fulfill this requirement, while August had 14. Tables listing the fine and coarse particulate matter concentrations data used in the evaluation are provided in Appendices A and B, respectively.

The mean, standard deviation, and minimum and maximum for each of the three months and the season are presented in Tables 4.3 and 4.4 for fine and coarse particulate matter concentrations, respectively.

Table 4.3 Statistical Evaluation Involving Fine Particulate Matter Concentrations.

Month	Mean		Std. Dev.		Minimum		Maximum	
	Obs.	Sim.	Obs.	Sim.	Obs.	Sim.	Obs.	Sim.
July	25.34	5.92	10.16	2.84	11.80	1.52	45.10	10.95
Aug.	24.63	5.89	10.65	2.95	10.57	1.55	53.33	10.90
Sept.	18.80	9.71	5.77	4.28	10.58	2.36	34.02	16.26
Summer*	22.71	7.20	6.74	3.32	12.65	1.80	39.86	12.48

* Three month mean, weighted by the total number of observations.

Table 4.4 Statistical Evaluation Involving Coarse Particulate Matter Concentrations.

Month	Mean		Std. Dev.		Minimum		Maximum	
	Obs.	Sim.	Obs.	Sim.	Obs.	Sim.	Obs.	Sim.
July	15.58	2.46	8.06	1.66	2.17	1.09	29.15	6.84
Aug.	14.54	2.06	8.17	1.17	4.67	1.04	33.43	5.46
Sept.	12.96	3.07	5.96	1.57	1.88	1.68	26.46	7.55
Summer	14.34	2.56	6.73	1.43	2.77	1.34	25.74	6.61

Examination of the tables reveals that in all cases, the model significantly underpredicted the fine and coarse concentrations. Scatter diagrams, which depict the correlation, or dependency of the simulated value (ordinate) upon the observed (abscissa) are presented in Figures 4.6 and 4.7. These too illustrate that the model simulations were significantly lower than the observed values. A line of best fit has been included as a reference. The correlation between the simulated and observed values of fine particulate matter was 0.533, indicating that 28.4% of the variance experienced by the observed values could be accounted for by the simulated values. Likewise, the correlation between the observed and simulated coarse

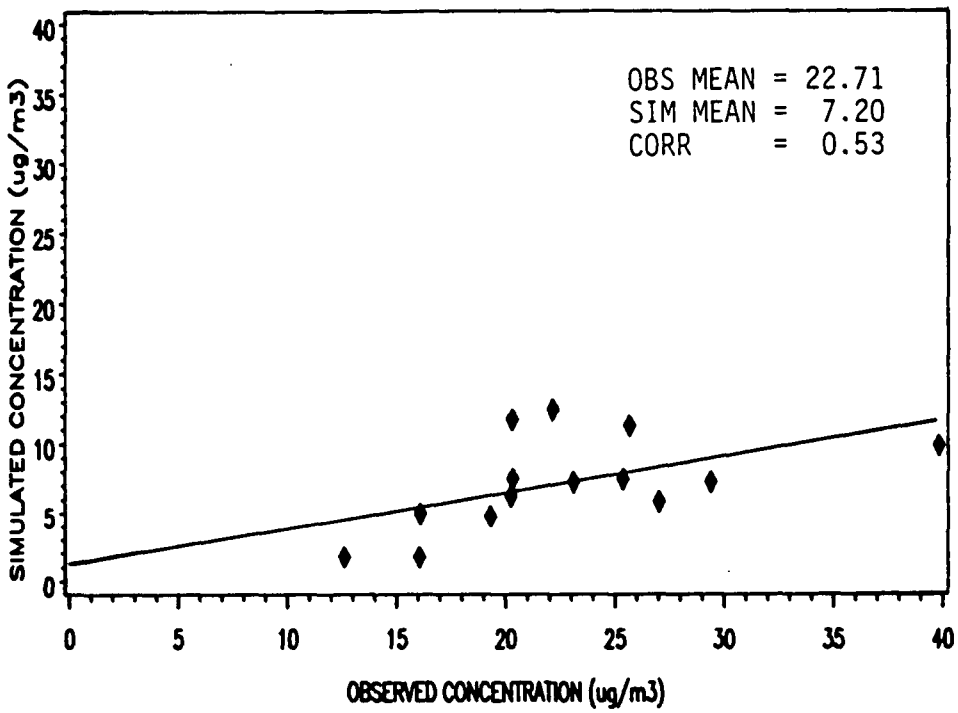


Figure 4.6 Scatter diagram of the observed vs. simulated fine particulate matter concentrations for the summer of 1980.

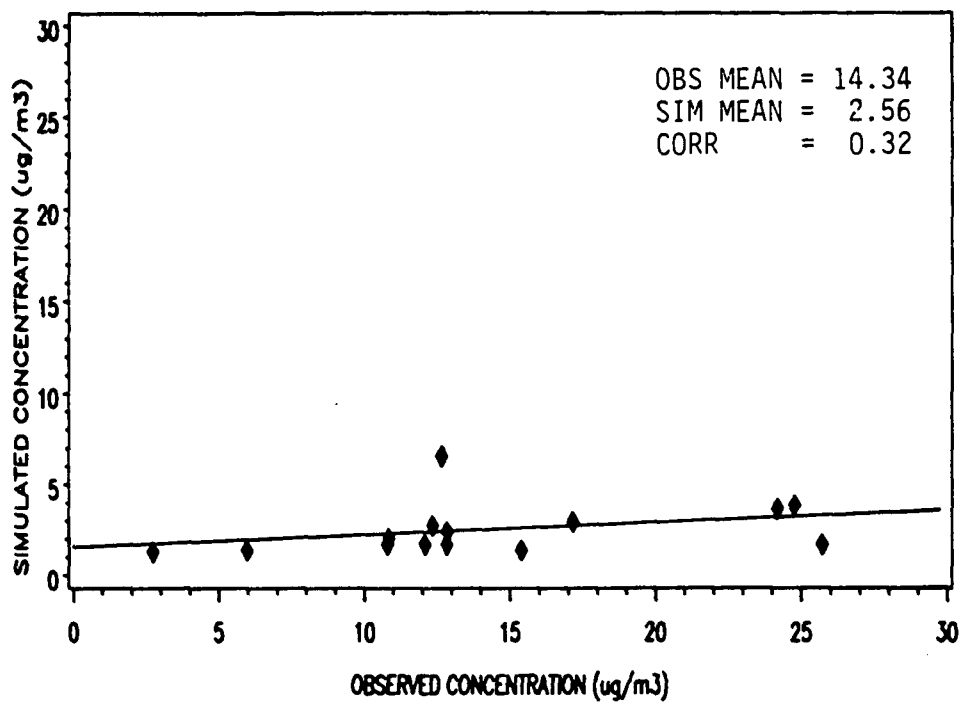


Figure 4.7 Scatter diagram of the observed vs. simulated coarse particulate matter concentrations for the summer of 1980.

concentrations was 0.322, indicating that 10.4% of the observed variance could be explained by the simulation.

The standard residuals ((observed-predicted)/observed) for each of the individual sites for the entire summer are depicted in Figures 4.8-4.9. These figures indicate that the model is consistent in its underprediction across the entire evaluation network. Standardized residuals range between 0.42 and 0.89 for the fine concentrations and between 0.48 and 0.93 for the coarse concentrations. This significant underprediction exhibited by the model is not surprising given the nature of the discrepancies discussed at the beginning of the section. All of the discrepancies inherent with the NAPAP emissions inventory would lend themselves to underpredictions by the model. Nearly 8% of the total TSP inventory was omitted because size fractions were not available. And even more significant is the exclusion of large emissions from open sources.

Several of the deficiencies inherent to the IPN would likewise result in the model underpredicting the concentrations. Designed primarily to characterize urban scale concentrations, the IPN had an overwhelming majority of its sites located within cities. In fact, of the 14 sites selected for this evaluation, 13 were designated as either center-city or suburban. Although regional influences dominate over local sources for fine particles, coarse particles would be adversely affected by such an arrangement. It is worth noting that the only remote site available for the study, RTP, actually showed fairly good agreement between the observed coarse concentrations (2.66 ug/m^3) and that simulated by the model (1.34 ug/m^3).

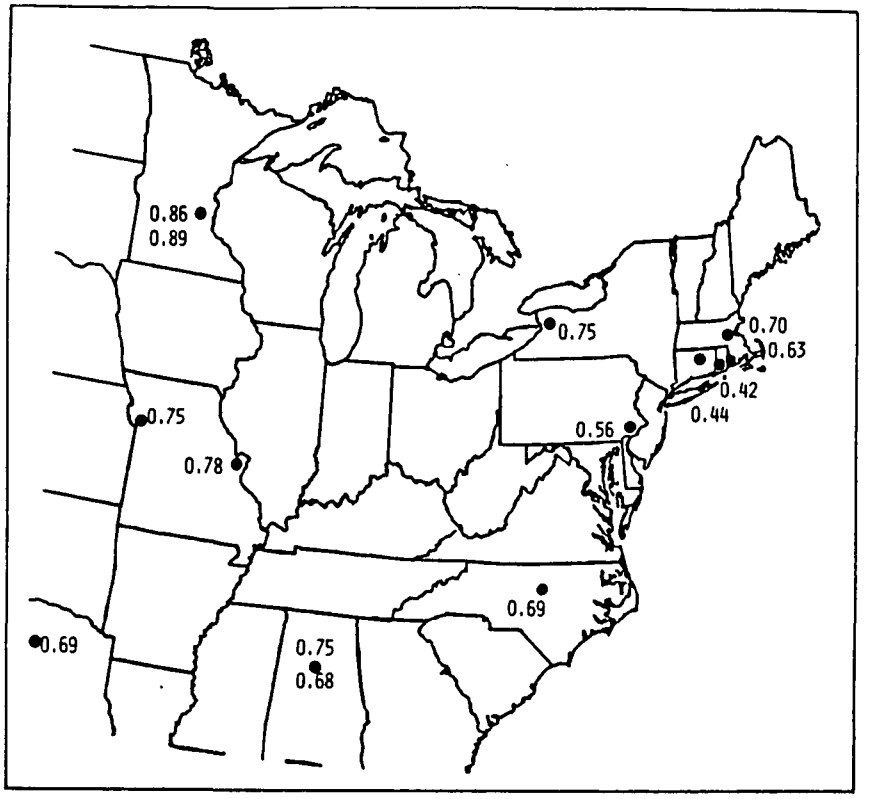


Figure 4.8 Standardized residuals ((O-P)/O) of the fine particulate matter concentrations for the summer of of 1980.

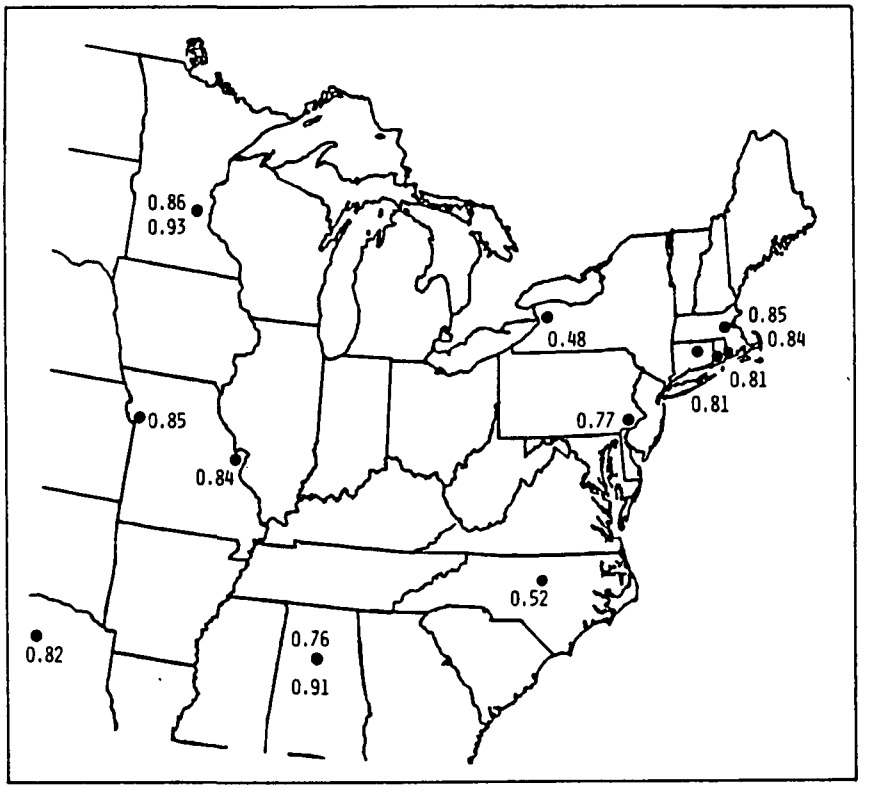


Figure 4.9 Standardized residuals ((O-P)/O) of the coarse particulate matter concentrations for the summer of 1980.

SECTION 5

CONCLUSIONS AND RECOMMENDATIONS

In response to the promulgation of the new, smaller, size discriminate National Ambient Air Quality Standards for IP, RELMAP has been modified to now include simple, linear parameterizations simulating the chemical and physical processes of fine and coarse particulate matter. Emphasis was placed upon the smaller particles for several reasons; first, the smaller sized particles were found to have a more adverse effect on health, and secondly, because the larger size particles had a large contribution from natural sources, attainment of federal standards was becoming more and more difficult.

Shifting the emphasis to the smaller particles enhances the utility of regional scale, Lagrangian models such as RELMAP. In this model, discrete puffs of SO_2, $SO_4^=$, fine and coarse particulate matter are subjected to linear transformation and wet and dry deposition processes as they are transported across the model's domain. RELMAP treats fine and coarse particulate matter as non-evolving pollutants and assumes physical or chemical transformation between the two to be negligible. RELMAP does however, consider the transformation of SO_2 into $SO_4^=$, which it treats as a function of solar insolation and moisture content. Dry deposition of SO_2, $SO_4^=$, and fine and coarse particulate

matter is treated as a function of land use, season, and stability. Wet deposition is treated by the model as a function of cloud type, pollutant concentration and precipitation rate.

Because these recently modified parameterizations are only accurate to a limited degree, they may be upgraded or even replaced in the future with more sophisticated parameterizations as further research is conducted. As an initial step in this possible refinement of RELMAP, the model was subjected to a sensitivity analysis. In this analysis, which employed actual emissions and meteorological data for July, 1980, variations found in the simulated concentrations of fine and coarse particulate matter, due to arbitrary +/- 50% variations from nominal values of the transformation rate and wet and dry deposition rates were examined.

Results of the analysis were recorded along a transect consisting of 15 grid cells which stretched across the model's domain. Each analysis consisted of two graphs, illustrating the absolute as well as the relative changes, with respect to a base case simulation. Simulated concentrations of fine and coarse particulate matter were found to be by far most sensitive to changes in the wet deposition rates of fine and coarse particulate matter, respectively. However, concentrations of fine particulate matter were quite insensitive to changes in the wet deposition rate of SO_2. Concentrations of coarse particulate matter were somewhat sensitive to dry deposition rates of coarse particles; however, fine particulate matter concentrations were less sensitive to dry deposition of fine particles and highly insensitive to dry deposition of SO_2. And finally, fine

particulate matter concentrations proved to be somewhat insensitive to the transformation rate of SO_2 into $SO_4^=$.

Future research should concentrate upon refining the parameterizations involving the wet deposition of both fine and coarse particulate matter. Not only has wet deposition proven to be the most influential parameterization employed by the model, it is also currently the least understood. Although the model proved to be somewhat less sensitive to the other parameterizations, future research should also address these areas as well, so that they too will parameterize the essential physical and chemical processes occurring in the atmosphere accurately.

In order to determine just how accurately these new parameterizations actually simulate the physical and chemical processes of the atmosphere, RELMAP was subjected to a model performance evaluation. The model was run for the summer of 1980, using actual meteorological data and emissions data from the NAPAP Version 5.0 emissions inventory. Simulations of ambient air concentrations of fine and coarse particulate matter were then compared to data from the IPN. Unfortunately, inadequacies inherent to both the emissions and validation data sets limited the scope of this evaluation.

As an illustration of these inadequacies, the NAPAP emissions inventory was designed primarily to support acid deposition modeling, not regional particulate modeling. Because of this, many deficiencies were found with the inventory, including the following: (1) most open source emissions were omitted from the

inventory (which by some estimates exceed 50,000 ktons of TSP), (2) the estimates of contributions from paved and non-paved roads, which account for 70% of the total inventory, are much lower in the NAPAP inventory than other independent estimates, (3) a total of 8% of the NAPAP inventory cannot be fractionalized, because particle size distributions are not available for many source classification codes.

The only way to alleviate these deficiencies is to reduce the tremendous amount of uncertainties in the estimates of the open source emissions. Such a solution may be forthcoming as the NAPAP Task Group II is scheduled to release, in the fall of 1987, a revised emissions inventory for open source emissions of TSP. Should this revised inventory include such major open sources of TSP as wind erosion, agricultural tilling, construction and mining operations, and should the new estimates include emissions from paved and non-paved roads which concur with other independent estimates, RELMAP'S accuracy and therefore its credibility as a regional-scale particulate model will improve.

A second major deficiency that proved to be detrimental to the model performance evaluation is the incompatibility of the IPN data. Of the 157 IPN sites that were operational at one time or another, only 14 were spatially and temporally compatible with the requirements of this evaluation. The IPN, like the NAPAP Version 5.0 emissions inventory, was not designed for regional scale particulate modeling. Rather the IPN was designed primarily to characterize the urban-scale concentrations of TSP, therefore, an overwhelming majority (144 of 157) of the sites were classified as either center city or suburban.

Another deficiency inherent to the IPN is the fact that observations were only recorded once every six days, resulting in a dearth of data. Since regional-scale pollution episodes have an average temporal span of several days, 24 h air concentrations sampled every sixth day are not be sufficient to capture the true variability of the ambient air concentration data. Therefore, when combined with the predominantly urban locations of the sites, the discontinuous sampling records of the IPN render the data inadequate for regional scale particulate modeling.

At the present time, there are no plans to implement a network that would fulfill the specific needs of regional scale particulate modeling. However, in the near future, a network proposed by NAPAP to assist in the evaluation of acid deposition models will begin monitoring pollutants on a regional scale at between 30 and 50 sites located in the eastern United States. If funded as proposed, continuous 12 h samples of fine particulate matter concentrations will be obtained, beginning in 1988, along with wet and dry deposition chemistry data. Although fine particulate matter is to be sampled, the analysis will concentrate on sulfate concentrations only. Therefore as currently proposed, the network fails to address the needs of regional scale particulate modeling.

Since appropriate data bases to evaluate regional scale particulate models do not exist, nor are any proposed, and because the cost of initiating and operating a network are prohibitive, Clark (1986) has recommended that the operational/analysis protocol of the proposed NAPAP network be

expanded to obtain an appropriate data base for evaluating regional scale particulate models. Because of its spatial and temporal distribution, the NAPAP network would provide an excellent data base. By supplementing the proposed network with fine and coarse particulate matter monitoring equipment, an appropriate data base can be generated for particulate modeling for a fraction of the cost needed to initiate and operate a new network.

Since the inadequacies discussed above have greatly limited the scope of this model evaluation, it must be considered preliminary at this time. Results of the performance evaluation indicate that RELMAP significantly underpredicted the average ambient concentrations of both fine and coarse particulate matter for the three month period. The observed and simulated fine particulate concentrations were 22.71 and 7.20 ug/m^3, respectively, while the observed and simulated coarse particulate concentrations were 14.34 and 2.56 ug/m^3, respectively. The correlation between the observed and simulated fine concentrations was 0.53, indicating that 28.4% of the variance was explained by the model. The correlation between the coarse simulated and observed concentrations was 0.32, indicating that 10.4% of the variance was explained.

Considering the nature of the deficiencies discussed above, such an underprediction by the model, though disappointing, is not surprising. Each of the deficiencies inherent to the NAPAP inventory and several inherent to the IPN data would indeed lend themselves to an underprediction by the model.

In order for RELMAP to become a credible regional

particulate model which can be used as a tool in assessing the effects of various emission control scenarios, it is critical that: (1) a revised TSP emissions inventory become available which more accurately emulates both the natural and anthropogenic emissions, and (2) adequate, regionally-representative, and continuous measurements of ambient air concentrations of both fine and coarse particulate matter be obtained.

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APPENDIX A

Table A.1 Fine Particulate Matter Concentrations (ug/m^3) for the Month of July.

	JULY					
STATION	02	08	14	20	26	AVERAGE
HUFFMAN	41.89	41.83	.	16.40	.	33.37
MTN. BROOK	36.05	33.60	37.34	10.59	13.63	26.24
HARTFORD	34.97	17.48	.	44.30	31.62	32.09
DOVER	.	.	.	.	.	.
BOSTON (Fire St)	.	.	14.17	50.95	29.23	31.45
BOSTON (S Cen)	.	17.42	13.25	48.63	29.46	27.19
MINNEAPOLIS (HS)	9.20	19.70	18.87	8.79	4.74	11.80
MINNEAPOLIS (N)	9.76	19.70	22.67	.	4.61	14.19
ST LOUIS	35.54	18.69	48.44	26.58	38.58	33.57
KANSAS CITY	14.04	10.83	7.98	.	17.46	12.58
BUFFALO	53.84	30.67	30.83	67.89	42.29	45.10
RTP	11.44	26.75	20.74	11.61	20.98	18.30
PHILADELPHIA	40.31	19.12	18.39	51.64	11.25	28.14
DALLAS	15.38	9.14	13.49	22.39	16.29	15.34

Table A.2 Fine Particulate Matter Concentrations (ug/m^3) for the Month of August.

	AUGUST						
STATION	01	07	13	19	25	31	AVERAGE
HUFFMAN	51.46	35.74	.	30.85	45.01	16.03	35.82
MTN. BROOK	34.65	29.31	20.85	20.07	31.98	9.76	24.44
HARTFORD	.	.	19.93	27.95	.	28.40	25.43
DOVER	26.88	15.81	37.32	16.36	21.19	9.17	21.12
BOSTON (Fire St)	54.28	21.04	30.26	14.33	40.05	19.56	29.92
BOSTON (S Cent)	48.38	20.39	21.62	11.59	21.66	.	24.73
MINNEAPOLIS (HS)	9.81	9.55	7.74	7.56	20.82	7.91	10.57
MINNEAPOLIS (Nic)	.	.	.	12.13	27.30	8.19	15.87
ST LOUIS	18.40	16.17	22.65	14.68	39.30	18.61	21.64
KANSAS CITY	.	.	10.61	.	19.68	10.72	13.67
BUFFALO	70.25	58.11	.	.	28.23	56.74	53.33
RTP	31.24	18.77	21.00	.	.	.	23.67
PHILADELPHIA	60.79	32.13	15.50	15.59	26.34	18.39	28.12
DALLAS	.	15.19	12.48	14.02	33.52	7.13	16.47

Table A.3 Fine Particulate Matter Concentrations (ug/m^3) for the Month of September.

	SEPTEMBER					
STATION	06	12	18	24	30	AVERAGE
HUFFMAN	29.61	.	14.08	23.66	6.61	18.49
MTN. BROOK	22.71	29.37	12.04	22.68	5.74	18.51
HARTFORD	12.68	20.61	7.20	6.71	14.35	12.31
DOVER	33.90	27.08	8.35	15.99	11.56	19.38
BOSTON (Fire St)	.	18.60	7.56	11.82	18.48	14.12
BOSTON (S Cen)	10.00	10.60	7.73	5.37	19.20	10.58
MINNEAPOLIS (HS)	11.49	23.30	.	.	19.89	18.23
MINNEAPOLIS (N)	11.82	29.17	7.50	8.74	31.56	17.76
ST LOUIS	23.59	.	.	.	.	.
KANSAS CITY	14.82	.	.	56.04	31.21	34.02
BUFFALO	25.75	18.65	13.88	15.57	45.35	23.84
RTP	17.11	26.70	16.79	.	.	20.20
PHILADELPHIA	32.36	27.41	13.58	9.40	18.98	20.35
DALLAS	11.04	27.96	22.31	12.59	8.94	16.57

Table A.4 Monthly and Seasonal Observed (Obs) and Simulated (Sim) Fine Particulate Matter Concentrations (ug/m^3) for the IPN Sites.

	JULY		AUGUST		SEPTEMBER		SUMMER	
STATION	OBS	SIM	OBS	SIM	OBS	SIM	OBS	SIM
HUFFMAN	33.37	5.98	35.82	5.86	18.49	10.09	29.43	7.28
MT. BROOK	26.24	5.98	24.44	5.86	18.51	10.09	23.15	7.28
HARTFORD	32.09	10.95	25.43	10.35	12.31	16.26	22.18	12.48
DOVER	.	.	21.12	9.21	19.38	14.46	20.33	11.79
BOS (Fire S)	31.45	5.70	29.92	5.46	14.12	11.57	25.41	7.53
BOS (S Cen)	27.19	5.70	24.73	5.46	10.58	11.57	20.37	7.53
MINN (HS)	11.80	1.52	10.57	1.55	18.23	2.36	12.65	1.80
MINN (Nic)	14.19	1.52	15.87	1.55	17.76	2.36	16.10	1.80
ST LOUIS	33.57	6.22	21.64	5.55	.	.	27.06	5.89
KANSAS CITY	12.58	4.07	13.67	3.22	34.02	7.25	19.34	4.82
BUFFALO	45.10	9.04	53.33	8.60	23.84	12.20	39.86	9.92
RTP	18.30	4.81	23.67	4.89	20.20	9.25	20.28	6.28
PHILADELPHIA	28.14	10.15	28.12	10.90	20.35	13.03	25.70	11.34
DALLAS	15.34	5.37	16.47	3.93	16.57	5.74	16.13	5.01

APPENDIX B

Table B.1 Coarse Particulate Matter Concentrations (ug/m^3) for the Month of July.

	JULY					
STATION	02	08	14	20	26	AVERAGE
HUFFMAN	17.05	41.94	.	28.48	.	29.15
MTN. BROOK	10.46	6.26	11.22	3.96	3.34	7.05
HARTFORD	10.52	14.12	.	15.50	8.67	12.20
DOVER	.	.	.	.	.	.
BOSTON (Fire S)	.	.	11.13	6.69	12.11	9.98
BOSTON (S Cen)	.	11.35	10.54	6.24	9.66	9.44
MINNEAPOLIS (HS)	15.06	13.17	24.13	7.80	6.26	13.28
MINNEAPOLIS (N)	17.54	19.06	27.63	.	12.00	19.06
ST LOUIS	36.68	19.63	26.71	18.50	21.76	24.66
KANSAS CITY	36.09	30.14	26.10	.	19.25	27.90
BUFFALO	30.22	9.53	19.66	8.72	10.73	15.77
RTP	1.00	3.15	1.52	3.09	0.91	1.93
PHILADELPHIA	20.15	14.55	13.71	10.07	9.79	13.65
DALLAS	15.86	12.57	20.17	28.09	14.59	18.26

Table B.2 Coarse Particulate Matter Concentrations (ug/m^3) for the Month of August.

	AUGUST						
STATION	01	07	13	19	25	31	AVERAGE
HUFFMAN	13.71	19.11	.	22.83	10.41	9.62	15.14
MTN. BROOK	3.91	11.23	8.56	7.88	4.94	2.66	6.53
HARTFORD	.	.	7.23	9.43	.	7.40	8.02
DOVER	16.29	13.43	10.82	5.28	10.82	7.80	10.74
BOSTON (Fire S)	12.59	17.75	13.22	7.94	13.32	16.57	13.57
BOSTON (S Cent)	8.90	14.82	12.34	6.29	11.27	.	10.73
MINNEAPOLIS (HS)	24.12	14.20	5.86	10.60	13.70	6.35	12.47
MINNEAPOLIS (Nic)	.	.	.	24.32	65.63	10.34	33.43
ST LOUIS	19.46	18.24	24.41	21.15	27.90	38.31	24.91
KANSAS CITY	.	.	26.97	.	22.30	26.50	25.25
BUFFALO	10.59	11.24	.	.	11.34	4.34	9.38
RTP	4.80	6.33	2.87	.	.	.	4.67
PHILADELPHIA	8.89	16.07	6.81	12.17	12.34	9.88	11.02
DALLAS	.	14.18	20.48	23.22	18.00	12.68	17.71

Table B.3 Coarse Particulate Matter Concentrations (ug/m^3) for the Month of September.

	SEPTEMBER					
STATION	06	12	18	24	30	AVERAGE
HUFFMAN	12.23	.	14.23	20.98	4.76	13.05
MT. BROOK	4.79	5.03	4.13	5.08	2.60	4.33
HARTFORD	11.10	16.82	4.81	9.93	14.94	11.52
DOVER	9.59	19.39	16.43	17.69	14.23	15.46
BOSTON (Fire S)	.	10.92	8.17	10.87	16.25	11.55
BOSTON (S Cent)	13.83	11.61	8.28	9.40	16.95	12.02
MINNEAPOLIS (HS)	14.73	9.79	.	.	14.27	12.93
MINNEAPOLIS (N)	22.61	24.33	18.23	26.46	40.67	26.46
ST LOUIS	38.31	25.91	.	.	.	.
KANSAS CITY	20.79	.	.	12.04	21.92	18.25
BUFFALO	7.38	12.39	11.72	20.88	11.26	12.73
RTP	0.82	2.89	1.94	.	.	1.88
PHILADELPHIA	7.89	20.75	4.70	10.85	19.73	12.78
DALLAS	11.59	14.14	26.45	18.64	6.97	15.59

Table B.4 Monthly and Seasonal Observed (Obs) and Simulated (Sim) Coarse Particulate Matter Concentrations (ug/m^3) for the IPN sites.

	JULY		AUGUST		SEPTEMBER		SUMMER	
STATION	OBS	SIM	OBS	SIM	OBS	SIM	OBS	SIM
HUFFMAN	29.15	1.54	15.14	1.04	13.05	1.68	15.42	1.42
MTN. BROOK	7.05	1.54	6.53	1.04	4.33	1.68	6.00	1.42
HARTFORD	12.20	1.64	8.02	1.71	11.52	2.91	10.87	2.08
DOVER	.	2.14	10.74	2.20	15.46	3.03	12.89	2.45
BOS (Fire S)	9.98	1.26	13.57	1.41	11.55	2.65	12.12	1.76
BOS (S Cen)	9.44	1.26	10.73	1.41	12.02	2.65	10.82	1.76
MINN (HS)	13.28	1.51	12.47	1.43	12.93	2.34	12.86	1.75
MINN (N)	19.06	1.51	33.43	1.43	26.46	2.34	25.74	1.75
ST LOUIS	24.66	4.06	24.91	2.89	.	4.81	24.80	3.91
KANSAS CITY	27.90	3.72	25.25	2.86	18.25	4.60	24.21	3.72
BUFFALO	15.77	6.84	9.38	5.46	12.72	7.55	12.70	6.61
RTP	1.93	1.09	4.67	1.13	1.88	1.81	2.66	1.34
PHILADELPHIA	13.65	2.62	11.02	2.44	12.78	3.39	12.39	2.81
DALLAS	18.26	3.36	17.71	2.34	15.59	3.30	17.19	3.00