United States
Environmental Protection
Agency

Atmospheric Sciences
Research Laboratory
Research Triangle Park NC 27711

Research and Development

EPA/600/S3-87/034 Jan. 1988

EPA

Project Summary

A Sensitivity Analysis and Preliminary Evaluation of RELMAP Involving Fine and Coarse Particulate Matter

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In response to the new, size discriminate federal standards for Inhalable Particulate 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 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.

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

Introduction

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 μm, to smaller, size discriminate Inhalable Particulate (IP) matter, which ranged in size from 0.0 to 15.0 μm. The IP was comprised of fine particulate matter (FINE-10), which included particles less than 2.5 μm 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 μm.

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 μm rather than a 15 μm criteria. Therefore, COARSE-15 was replaced by COARSE-10, which included particles greater than or equal to 2.5 μm, but less than or equal to 10.0 μm.

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.

Model Background

RELMAP is a mass-conserving, regional-scale Lagrangian model that performs simulations over 1° by 1° grid cells covering the eastern two-thirds of the United States and southeastern Canada. 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. 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.

Because 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, the model simulates both horizontal and vertical diffusion through simple parameterizations. RELMAP divides the atmospheric boundary layer into three layers. The first layer is between the surface and 200 m, and 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.

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 layers into which they are emitted. 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.

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.

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

Dry deposition of SO_2, $SO_4^=$, fine and coarse particulate matter is a highly variable, complex process that is parameterized by RELMAP as a function of land use, season, and stability index. Twelve land use categories, categorized by surface characteristics and vegetation type were gridded to RELMAP's 1° by 1° domain. Typical dry deposition velocities used in the model 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 parameterizations discussed 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.

Because coarse particulate matter consists of a wide range of particle diameters, two sets of dry deposition velocities are used by the model. The first set applies to particulate matter with diameters of 5 μm, and the second set applies to particles with diameters of 10 μm. The dry deposition velocities for particles with a 5 μm diameter range between 0.4 and 5.0 cm/s, and between 1.0 and 6.0 cm/s for the 10 μm diameter particles. Unlike SO_2, $SO_4^=$, and fine particulate matter, the dry deposition velocities of coarse particulate matter are much less dependent of the time of day or the season; therefore, diurnal and seasonal variations are considered negligible.

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 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.

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 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. 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 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. Results of the sensitivity analysis were recorded along a specific transect that stretched across the model's domain from Alabama to Quebec. The sensitivity analysis of coarse particulate matter revealed 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, decreasing either the wet or dry deposition of coarse particulate matter results in increased concentrations. For a 50% decrease in the wet deposition, the concentration increases an average of 30 to 50%, but 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. 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%.

Analysis of 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 $SO_4^=$, reveals many of the same characteristics as noted with the coarse particulate matter. As expected, the wet deposition of fine particulate matter had the largest impact upon the concentration field. 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. The influence of SO_2 wet deposition, however, proves to be minimal.

Analysis of the dry deposition for both SO_2 and fine particulate matter, reveals that the influence of SO_2 dry deposition on the concentration of fine particulate matter proved to be all but non-existent. 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. 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, the sensitivity of fine particulate matter concentration to changes in the transformation rate is somewhat 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.

Preliminary Model Performance Evaluation

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. 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. Although Version 5.0 of the 1980 NAPAP Emissions Inventory represents by far the most comprehensive and highest quality emissions data set available, it was developed 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 with 46,560 ktons or 62.8% of the total NAPAP TSP inventory being emitted within the model's domain. Of this total, 90.23% can be attributed to area sources, and 9.77% can be attributed to point sources. Of the 4,550 ktons of TSP attributed to point sources, 14.20% are emitted as particles with diameters larger than 10 μm, 7.56% are emitted as fine particles, 7.29% are emitted as coarse particles, and 70.95% cannot be fractionalized. Fractionalization of the area source emissions 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 μm, and 0.85% cannot be fractionalized.

These non-fractionalized percentages illustrates one of the major deficiencies of the NAPAP TSP inventory. A large percentage of the many point and area source categories designated by NAPAP do not have particle size distributions. Because of this, more than 3,584 ktons or roughly 7.7% of the TSP emitted from point and area sources located within the model's domain cannot be fractionalized, or broken down into the respective size categories. 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 TSP inventory is the omission of many of the "open" source emissions, which are described as sources of air pollution too great in extent to be controlled by enclosure. Open source emissions, which are extremely difficult to estimate, are equivalent in magnitude, under the most conservative of estimates, to the anthropogenic sources — yet most are excluded from the inventory. Examples of open sources of TSP excluded from the inventory include: agricultural tilling, wind erosion, construction activity, and mining operations. Estimates of those open sources which are included in the NAPAP inventory are often much lower than other independent estimates. For example, independent estimates of TSP emissions from paved and non-paved roads, which account for over 70% of the TSP emissions in the NAPAP inventory, are more than an order of magnitude higher than the NAPAP estimates.

Because of the number and seriousness of these deficiencies, any model performance evaluation using the NAPAP inventory as a source of TSP 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.

In order to adequately evaluate a regional scale model such as RELMAP, which has a 1° by 1° 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 Inhalable Particulate Network (IPN), which was developed and implemented by the Environmental Monitoring Systems Laboratory (EMSL) in conjunction with the Office of Air Quality Planning and Standards (OAQPS), was primarily designed to characterize urban scale concentrations of suspended particulate matter, since the attainment of air quality standards is evaluated over this scale. 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.

The IPN became operational during April 1979, when 57 sites located throughout the United States went online using hi-vol, dichotomous and size selective inlet samplers to collect data, and eventually grew to 157 sites in 1981. 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. 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.

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). When combined with the amount of "down time" experienced at each site, these sixth-day observations resulted in a dearth of data, which in turn made the model evaluation very difficult and preliminary at best. 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.

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. Monthly and seasonal simulated values of fine and coarse concentrations (expressed in $\mu g/m^3$) were compared to the 14 compatible sites from the IPN. 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 concentrations was 0.322, indicating that 10.4% of the variance could be explained by the simulation.

The standard residuals ((observed-predicted)/observed) for each of the individual sites for the entire summer indicate that the model consistently underpredicts 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 throughout this section.

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. Shifting the emphasis to the smaller particles enhances the utility of regional scale, Lagrangian models such as RELMAP. 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.

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 sensitive 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.

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, in which simulations of ambient air concentrations of fine and coarse particulate matter were compared to data from the IPN. Unfortunately, inadequacies inherent to both the emissions and monitoring data sets limited the extent of this evaluation.

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 no-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 the major open sources of TSP, RELMAP's accuracy and therefore its credibility as a regional-scale particulate model will improve.

A second major deficiency that also proved to be detrimental to the model performance evaluation is the incompatibility of the IPN data. Like the NAPAP Version 5.0 emissions inventory, the IPN was not designed for regional scale particulate modeling. Because of this, many obstacles were encountered when trying to evaluate the model with the IPN data, including the following: (1) designed primarily to characterize the urban-scale concentrations of TSP, an overwhelming majority (144 of 157) of the IPN sites were classified as either center city or suburban; (2) observations were only recorded once every sixth day (most sites also had incomplete records) resulting in a dearth of data; (3) many of the monitoring sites did not come "online" until after the evaluation period; and (4) many of the sites were located outside the model's domain. The combination of these deficiencies render the data inadequate for evaluating regional scale particulate models.

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. 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, it is 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.

Unfortunately, the inadequacies discussed above have greatly limited the scope of this model evaluation, therefore, 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 concentrations were 22.71 and 7.20 $\mu g/m^3$, respectively, while the observed and simulated coarse concentrations were 14.34 and 2.56 $\mu g/m^3$, respectively. The correlation between the observed and simulated fine and coarse concentrations were 0.53 and 0.32, respectively. Considering the nature of the deficiencies discussed above, such an underprediction by the model, through 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 adjustment scenarios, it is critical that: (1) a revised TSP emissions inventory become available which more accurately emulates both the natural and anthropogenic emissions of TSP, and (2) adequate, regionally-representative and continuous measurements of ambient air concentrations of both fine and coarse particulate matter are obtained.

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The complete report, entitled "A Sensitivity Analysis and Preliminary Evaluation of RELMAP Involving Fine and Coarse Particulate Matter," (Order No. PB 88-114 012/AS; Cost: $14.95, subject to change) will be available only from: only from:

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