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Project Summary

Evaluation of Short-Term NO_2 Plume Models for Point Sources: Volumes 1 and 2

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Four mathematical models for atmospheric pollution transport and reactivity were tested on MISTT data base for their ability to predict patterns of ambient NO_2 concentrations in the vicinity of large point sources. The four models tested were: (1) the Reactive Plume Model (RPM-II), (2) the Total Conversion Method (TCM), (3) the Oxidant Limiting Method (OLM), and (4) the Conservation of NO_x and Oxidant Method (CNOM). Of these methods, RPM-II was by far the most sophisticated, and yielded more accurate predictions than the other three models.

This Project Summary was developed by EPA's Environmental 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 order information at back).

Introduction

The burning of fossil fuels by automobiles, power plants, and refineries produces appreciable amounts of oxides of nitrogen (NO_x), which are then released into the atmosphere. These emissions consist primarily of relatively harmless nitric oxide (NO); however, under suitable conditions, such as in the presence of reactive hydrocarbons and sunlight, NO can be oxidized to form nitrogen dioxide (NO_2).

Earlier epidemiological studies indicated that long-term exposure to high ambient NO_2 concentrations was associated with a higher incidence of respiratory disease. In addition, there has been some recent concern about the effects of short-term peak NO_2 concentrations on the frequency and severity of acute respiratory disease. The results of some studies show that the magnitude of the short-term peak NO_2 concentration had a significantly greater influence than either the duration or frequency of exposure. Because of the importance of these results, the U.S. Environmental Protection Agency (EPA) is giving serious consideration to promulgating short-term NO_2 air quality regulations. Part of this consideration is the review of techniques that can be used to predict short-term (about 1 h) average, ground-level NO_2 concentrations. Specific attention is being given to the ability to predict NO_2 impacts due to NO_x emissions from large point sources (e.g., power plants and other large combustion processes).

Under the sponsorship of EPA, Systems Applications, Inc. (SAI) has selected several models that can be used to predict ground-level NO_2 concentration impacts from point sources and has applied the selected models to MISTT data base (a large aerometric and meteorological data base collected near St. Louis) for the purpose of evaluating their performance. The MISTT data were selected by EPA because the

MISTT program involved a large urban and point source monitoring effort that included a significant number of aircraft plume measurements.

Description of Models

The models evaluated in this study were: (1) the Reactive Plume Model (RPM-II); (2) the Total Conversion Method (TCM); (3) the Oxidant Limiting Method (OLM); and (4) the Conservation of NO_x and Oxidant Method (CNOM). The last three are very simple methodologies that have been suggested for use with standard Gaussian point source dispersion models. RPM-II, on the other hand, is much more sophisticated, allowing treatment of time-varying winds, dispersion rates, reaction rates, and can accommodate the use of very large photochemical kinetic mechanisms These models were chosen to provide a range of complexity in NO_2 predictions, from the very simplest approach (TCM) to one of the more advanced reactive plume models (RPM-II).

In the Conservation of NO_x and oxidant method, the equilibrium relationship among NO, NO_2 and ozone (O_3) is used in conjunction with the assumptions of oxidant and NO_x conservation to simplify the chemical kinetics. CNOM suffers the limitation that beyond a few kilometers from the source its simplified treatment of chemistry is invalid in hydrocarbon-rich environments. In addition, any deficiencies in the dispersion parameter computations—the Gaussian dispersion relationship, for example, has many known limitations—will be reflected in predicted NO_2 concentrations.

An even simpler model is the oxidant limiting method (OLM). The OLM assumes that NO_2 concentrations in the plume are equal to the background O_3 plus some small fraction of the emitted NO concentration. The dependence on the background O_3 is an acknowledgment that one NO_2 molecule is formed for every reaction of O_3 and NO molecules and that NO_2 thus may be limited by the background O_3 in an NO-rich plume. However, it does not explicitly account for diffusion, entrainment, or the photostationary relationship. Another simplification, called total or partial conversion (TCM or PCM), assumes that all or some fraction of the emitted NO_x is ultimately converted to NO_2.

The TCM and OLM suffer the same deficiencies as CNOM; moreover, these methods are further limited by their own, more restrictive, assumptions. However, in many cases they are useful screening procedures. TCM and OLM can be used quickly and inexpensively to obtain an estimate of the magnitude of the potential NO_2 impacts.

RPM-II considers an array of well mixed reactors or cells across the plume, each of which moves downwind and disperses along the mean-wind trajectory. The model can accommodate temporally and spatially varying wind speeds, reaction rates, ambient concentration levels, and dispersion or entrainment rates. As currently configured, RPM-II uses a state-of-the-art 68-step carbon bond photochemical reaction mechanism developed by Whitten and Hogo*; other mechanisms can be easily substituted for the carbon bond mechanism if desired. The most severe limitation of an involved reactive plume model like RPM-II is the requirement for valid ambient concentration estimates of reactants along the plume trajectory. These must by measured, predicted, or hypothesized. Ambient hydrocarbons are extremely difficult to characterize even by measurement, yet predictions of plume NO_2 are very sensitive to hydrocarbons. Like most other dispersion models, RPM-II is also limited by the user's ability to properly specify wind speeds and dispersion rates, especially in complex terrain applications. But despite such limitations, reactive plume models like RPM-II are clearly more realistic in their approach to NO_2 estimation than are CNOM, OLM, or TCM.

Data Base

The data required for the application of the models selected are:

- Atmospheric stability, dispersion coefficient, or horizontal plume spread as a function of downwind distance and time.
- Point source emissions rates for NO, NO_2, sulfur dioxide (SO_2), sulfates, and hydrocarbons.
- Source data:
  - Plume heights.
  - Source parameters: flow rates, stack temperatures.
  - Source locations.
- Ambient concentrations for all species as functions of location and time.
- Sunlight intensity as a function o location and time.
- Inversion heights or vertical plume spreads as functions of location and time.
- Wind field.

These data will also satisfy all requirements of both RPM-II and the three simple NO_2 estimation procedures.

At the start of this study, the usefulness of the MISTT data base for applying and evaluating NO_2 prediction techniques was uncertain, because the data base was designed to characterize plume sulfur and aerosol transport and transformations, not to support reactive plume modeling. However, a significant amount of plume data was collected during the MISTT study and much of it for a large point source (Labadie power plant plume). This study focused on the Labadie power plant plume data collected during the MISTT program. Most of the required data were available as part of the MISTT data base or from other data sources. However, after careful examination of the MISTT data, the airborne hydrocarbon concentration measurements were found invalid. Plume dispersion parameters were usually difficult to estimate with much certainty from the airborne concentration profiles.

To compensate for uncertainties in the dispersion data, the input dispersion rates were adjusted so that the following equation was approximately satisfied:

$$A(S) = [(E_i/U(S) - (C_{ia}A_o)]/(C_{ip}(S) - C_{ia})$$

where

- $A(S)$ = plume cross-section area at downwind distance S
- E_i = emissions rate of species i (NO_x or SO_2)
- $U(S)$ = wind speed at distance S
- C_{ia} = measured ambient concentration of species i
- A_o = exit area of stacks (121.2 m^2)
- $C_{ip}(S)$ = measured average plume concentration of species i at distance S

Adjustments were made to the total ambient hydrocarbon levels so that the plume O_3 concentrations predicted by RPM-II approximately matched the measured values. The reactivity of the hydrocarbon mass (i.e., the species distribution of hydrocarbons) was determined from hydrocarbon analyses carried out for the RAPS program (a ground-level monitoring program conducted in the same area during the MISTT program.)

These adjustments to the input data preclude a strict evaluation of model

*Whitten, G Z and H Hogo, Mathematical Modeling of Simulated Photochemical Smog. EPA-600/3-77-011, U.S. Environmental Protection Agency, Research Triangle Park, NC, 1977, 307 pp.

performances or verification of model predictions. However, it is possible to judge whether the models are capable of acceptable performance when reasonable input values are selected.

In addition, a "restricted" data base was assembled for input to the models. The purpose of using this data set was to evaluate model performance for the limited data normally available to a prospective user. Model input values for all parameters in the "restricted" data set were identical to those in the "unrestricted" set, except for wind speeds, plume widths, and plume depths. These exceptions were instead derived from local National Weather Service station data and standard Pasquill-Gifford-type dispersion curves.

A compendium of data collected by Meteorology Research, Inc. and the University of Washington during the MISTT program is presented in plotted form in Volume 2 of the Project Report. This includes maps showing aircraft flight paths and graphs of the data collected along the paths.

Results

Ten case-study days were selected from the MISTT study data base, and 28 observations of average plume concentrations at various downwind distances were derived from the data on those 10 days. The results of the model runs with the "unrestricted" and "restricted" data bases were studied with the aid of several statistical and graphic techniques. The RPM-II performed better than the simple NO_2 models in both the "restricted" and "unrestricted" cases. This result might be expected because RPM-II is intended to account in greater detail for more processes than are the simpler models.

Inspection of scatter diagrams reveals that RPM-II is capable of matching the observations more closely than the simple models across a wide range of NO_2 concentrations. The least-squares best fit line has a slope of 0.887 and an intercept of 1.6 ppb, compared with an ideal slope of 1.0. The RPM-II clearly performed within the expected accuracy of the monitoring instruments ($\sim \pm 15$ percent of the true value). The computed statistics are also good, but they do not constitute validation of RPM-II performance because of the input data uncertainties. Rather, the following conclusions about RPM-II are warranted:

- The model is capable of acceptable performance when reasonable ambient hydrocarbon and plume dispersion input values are selected.
- No apparent defect that affects performance is revealed in the model's formulation, although the formulation does contain several simplifying assumptions.

Because the simple models contain a relatively simplistic treatment of chemistry, they may not be appropriate for use in situations where hydrocarbon oxidation significantly influences NO_2 concentrations. It is therefore desirable to define concisely the character of the situations requiring more sophisticated models. On the basis of the simulations carried out in this study, however, it is clear that several important parameters interact to determine NO_2 production rates. Thus, a concise definition of conditions requiring a sophisticated model is not possible on the basis of these results. NO_2 production appears generally to depend on the existence of a sufficient amount of ambient ozone and hydrocarbons, solar insolation, and plume dispersion in combination. The precise combinations depend on the following variables:

- Amount of NO_x present in the plume and in the ambient air.
- Time of release from the stack.
- Ratios of NO to NO_2 in the plume and in the ambient air.
- Wind speed and variability.
- Split of the reactive hydrocarbons.

Conclusions

The four models, RPM-II, TCM, OLM, and CNOM performed reasonably well by comparison with MISTT data both "unrestricted" and "restricted" input data sets. Of the four, RPM-II was the superior performer using both data sets over a wide range of concentration levels. TCM, OLM, and CNOM tended to underpredict but mostly at the high concentration ranges. It must be stressed that model performances in this study cannot be construed as a verification of model performances because of input data deficiencies and the input data adjustments that were necessary.

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The complete report consists of two volumes entitled "Evaluation of Short-Term NO_2 Plume Models for Point Sources:"

"Volume I. Technical Discussion," (Order No. PB 82-234 329; Cost: $17.50, subject to change).

"Volume II. Data," (Order No. PB 83-217 232; Cost: $4.50, microfiche only, subject to change).

The above reports will be available only from:

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