Air Pollution Modeling and Its Application IV

Edited by
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PREFACE

In 1969 the North Atlantic Treaty Organization established the Committee on the Challenges of Modern Society. Air Pollution was from the start one of the priority problems under study within the framework of the pilot studies undertaken by this Committee. The organization of a yearly symposium dealing with air pollution modeling and its application is one of the main activities within the pilot study in relation to air pollution.

After being organized for five years by the United States and for five years by the Federal Republic of Germany, Belgium, represented by the Prime Minister's Office for science Policy, became responsible in 1980 for the organization of this symposium.

This volume contains the papers presented at the 14th International Technical Meeting on Air Pollution Modeling and its Application held in Copenhagen, Denmark, from 27th to 30th September 1983. This meeting was jointly organized by the Prime Minister's Office for Science Policy, Belgium, and the National Agency of Environmental Protection, Air Pollution Laboratory, Risø National Laboratory, Denmark. The conference was attended by 103 participants and 43 papers have been presented. The members of the selection committee of the 14th I.T.M. were A. Berger (Chairman, Belgium), W. Klug (Federal Republic of Germany), K. Demerjian (United States of America), L. Santomauro (Italy), H. Van Dop (The Netherlands), H.E. Turner (Canada), C. De Wispelaere (Coordinator, Belgium).

The main topic of this 14th I.T.M. was Parametrization of Transformation and Removal Processes in Air Quality Modeling. On this topic a review paper was presented by Ø. Hov (Norwegian Institute for Air Research). Other topics of the conference were : Coastal Meteorology related to Air Pollution Modeling, Lagrangian Modeling for Synoptic Range Transport, Atmospheric Experiments pertinent to Air Quality Modeling, Practical Applications of Air Quality Modeling.

U. Peching (Department Meterology, San Jose State University, California, U.S.A.) and F.L. Ludwig (SRI International, Menlo Park, U.S.A.) presented a review paper as an introduction to the topic on Costal Meteorology related to Air Pollution Modeling Dr. J. Knox (Lawrence Livermore National Laboratory, California, U.S.A.) gave a review on the topic Lagrangian Modeling for Synoptic Range Transport.

On behalf of the selection committee and as organizer and editor I should like to record my gratitude to all participants who made the meeting so stimulating and the book possible. Among them I particulary mentíon the chairmen and rapporteurs of the different sessions. Thanks also to the local organizing committee, chaired by Dr. J. Fenger. Thanks also to Mrs. J. Husted and P. Sondergaard, who were the Conference Secretaries. Finally I have the pleasure to record my thanks to Mrs. N. Desees and L. Mille and Miss J. Bertrand for typing and to Mr. D. Poelman for preparing the papers.

C. De Wispelaere

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1: PARAMETERIZATION OF TRANSFORMATION AND REMOVAL PROCESSES IN AIR QUALITY MODELING

Chairmen: A. Berger
Ø. Hov
U. Pechinger

Rapporteurs: J. Van Ham
E. Lyck
P. Zannetti

ASPECTS OF THE PARAMETERIZATION OF TRANSFORMATION AND REMOVAL PROCESSES IN AIR QUALITY MODELLING

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1 INTRODUCTION

The air quality is linked to the ambient air concentration of species like ozone (O_3), nitrogen dioxide (NO_2), peroxyacetylnitrate (PAN), aldehydes, carbon monoxide (CO), sulphur dioxide (SO_2), sulphate ($SO_4^=$), nitric acid/nitrate (HNO_3/NO_3^-), soot and many other types of particulate material (containing trace metals, hydrocarbons, chlorinated hydrocarbons etc.). Ozone alone, or in combination with SO_2 or NO_2, is responsible for up to 90% of the crop losses in the U.S. caused by air pollution. An estimate made for the U.S., assuming that all areas just met the current O_3 standard of 120 ppb as hourly average, showed a loss of 2 to 4% of the crop production. A test program, utilizing field chambers where the ozone concentration could be controlled, demonstrated yield reductions in all crops at seasonal 7h/day mean O_3 concentrations of 60 - 70 ppb when compared with a control value of 25 ppb, thought to represent the natural background (Heck et al., 1982).

At the 1982 Stockholm conference on acidification of the environment it was stated in a consensus report that "the recently reported forest damage in an estimated one million hectares of Central Europe seems to be related to (among others) the direct effects of gaseous pollutants and soil impoverishment, and toxicity arising from very large amounts of wet and dry deposition" (Hileman, 1983). For a long time the most noticeable effect from acid rain has been the lowering of pH in thousands of lakes in Scandinavia and eastern North America. A consequence has been a substantial increase in the amount of dissolved aluminium, which has caused a wide spread decimation of the fish population.

Pollutants affecting the air quality are emitted directly or they are derived chiefly from the emissions of nitrogen oxides (NO_x), SO_2 and hydrocarbons (HC). An attempt at characterizing in terms of spatial and temporal scales of dispersion and transformation the most important chemical species which affect the air quality, is shown in Table 1. If the atmospheric fate of a given pollutant is to be computed by a model, it is required that the temporal and spatial domains are at least as indicated in Table 1. If an attempt is made at analysing e.g. ozone generation by constructing a model covering an area extending a few km and over a few hours, it is likely that the computational result is strongly dependent on the boundary conditions.

The chemical species which affect the air quality through their presence in the atmospheric boundary layer (ABL), are in general removed from the ABL on a time scale comparable to the typical time length between ABL break-down situations. At our latitudes, the ABL typically takes 2 - 4 d to reach a break-down situation (Smith and Carson, 1977). Break-down may occur as a result of large scale convective instability where convective elements may carry significant amounts of heat, moisture and pollution upwards into layers otherwise not affected by the underlying surface. Break-down may occur at fronts through upsliding motion, in mountainous regions where the vertical mixing may be considerable, and by continuous synoptic-scale vertical motion in combination with the diurnal cycle in the depth of the boundary layer which causes the contents of the boundary layer to be pumped upwards in the middle of the day, where it is left behind to be acted upon by the steady synoptic upward motions. Water soluble species may be rained out by precipitation associated with ABL break-down. The ABL vertical pumping effect distributes species like N_2O, CFC's, CH_4, O_3, CO and others into the free troposphere, to be acted upon on a global spatial scale.

The design of a particular air quality model has to be chosen on the basis of the problem to be analysed. The calculation of street canyon concentrations of a primary pollutant like CO requires different chemical and physical/meteorological considerations compared to the modelling of the generation, transport and loss of ozone or sulphate in the ABL. The time between ABL break-downs defines important temporal and spatial scales for ABL air quality modelling, however, and in this paper emphasis will be put on chemical transformation and removal of HC, SO_2 and NO_x over several days.

It is to be understood that parametrization of atmospheric transformation and removal processes of chemical species involves assigning numerical values to the significance of the known pathways for transformation and removal. In a broader sense, parametrization also includes the mathematical framework in which the numerical values for physical and chemical processes are to be embedded, and how to solve the mathematical equations which arise.

Table 1. Characterization in terms of spatial and temporal scales of dispersion and transformation of chemical species affecting the air quality.

Species	Scales for dispersion and transformation		Comments
	Space	Time	
O_3	$\leq$ 1000 km	$\leq$ 10 d	
PAN	$\leq$ 1000 km	$\leq$ 5 d	
SO_2	$\leq$ some 100 km	$\leq$ 2 d	
$SO_4^=$	$\leq$ 1000 km	$\leq$ 5 d	
NO_2	$\leq$ 1 km	$\leq$ 1 h	Scales appropriate for traffic. NO_2 may also be a dominating oxidized nitrogen species in long-range transported air (Grennfelt, 1979).
HNO_3 (NO_3^-)	$\leq$ 1000 km	$\leq$ 10 d	
CO	$\leq$ 1 km	$\leq$ 1 h	Scales appropriate for traffic. There is also a significant anthropogenic influence on the tropospheric global CO budget.
HCHO (aldehydes)	$\leq$ 10 km	$\leq$ 10 h	
C_6H_6 (benzene)	$\leq$ 1000 km	$\leq$ 10 d	
N_2O, CH_4, CFC, CO_2, O_3	global	years	Anthropogenic influence on the global tropospheric budgets may affect the future global climate.

2 MATHEMATICAL FORMUALTION OF AN AIR QUALITY MODEL

Mass conservation of each chemical species is a common requirement in air quality modelling. This can be written as

$$\frac{\partial c_i}{\partial t} + \nabla \cdot \vec{v} c_i = \nabla \cdot D_i \nabla c_i + R_i \qquad (1)$$

where c_i denotes the concentration of species i, $\vec{v}$ is the wind field (stochastic variable), D_i molecular diffusion coefficient for species i, while R_i includes the effects of atmospheric transformation, removal and emission. In most air quality models the ensemble averaged atmospheric diffusion equation is applied

$$\frac{\partial \langle c_i \rangle}{\partial t} + \nabla \cdot \bar{\vec{v}} \langle c_i \rangle = \nabla \cdot (K \cdot \nabla \langle c_i \rangle) + R_i(\langle c_1 \rangle, \ldots, \langle c_p \rangle),$$

$$i = 1, \ldots, p \qquad (2)$$

where $\langle c_i \rangle$ is the ensemble averaged concentration (the brackets will be omitted in the following), $\bar{\vec{v}}$ is the deterministic wind velocity and K the tensor introduced to approximate the turbulent transport term by the mean concentration gradient, p is the number of components (see e.g. Seinfeld, 1975, McRae et al., 1982). In this paper the R_i in eq. (2) will be examined, since this term describes the effects of chemistry, removal and emission on the time development of the concentration of species no. i.

In Figure 1 is shown a diagram of the factors which make up R_i. Eq. (2) is often taken as the general form of the continuity equation for a chemical species. In practical applications, less general forms of the equation are used depending on the problem to be analysed, by reducing the dimensionality to two or one, by transformation into a trajectory model, etc. Recent reviews are published by McRae et al., 1982, and Johnson, 1983. In practical applications, the mathematical formulation is a compromise based on many factors: The quality and detail of the meteorological data, emission fields, data describing chemical transformations, availability of computational and economical resources, and availability of air quality data against which the model can be compared.

The numerical solution of eq. (2) or some simplified form of eq. (2) is not straight forward. The component of the equation system which describes the chemical transformation is in general non linear, and exhibits stiff properties. This means that there is a wide range of chemical decay times involved. Special techniques have to be applied to keep the computer cost down and obtain

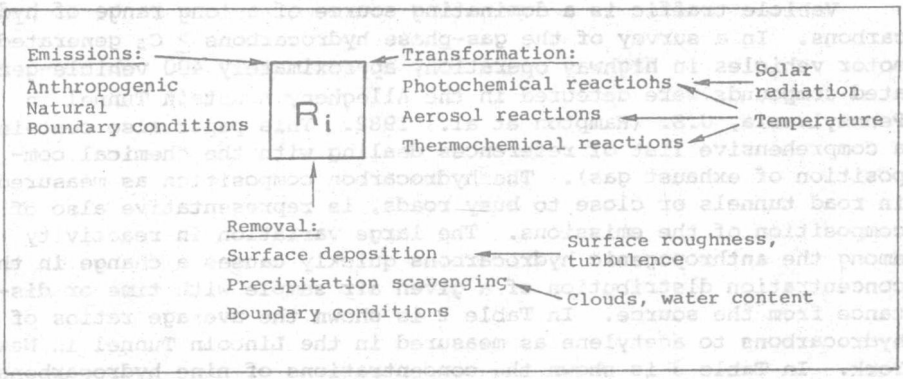

Figure 1. Factors in the term R_i in eq. (2).

solutions with acceptable numerical accuracy (Gear, 1971, Whitten, 1976). A so called quasi-steady state approximation technique has been developed by Hesstvedt et al., 1978, which cuts down the computer cost for the integration of kinetic equations, but which depends on other methods (e.g. Gear, 1971) for cross checks to assess the numerical accuracy (see also Hov et al., 1978a and Derwent and Hov, 1979).

Recently several workers have applied operator splitting techniques to solve particular versions of eq. (2). It is then possible to apply special numerical procedures to solve the various parts of the equation, where the basic elements are those due to transport and those due to chemistry (see McRae et al., 1982, Hov, 1983a).

3 PARAMETRIZATION OF CHEMICAL TRANSFORMATIONS IN THE GAS PHASE

3.1 Composition of atmospheric hydrocarbons

The emissions of organic material into the atmosphere have an exceedingly complex composition. Even though the mass of natural emissions dominates on a global basis, it is generally believed that natural organic emissions play a minor role in the formation of pollutants which affect the air quality (Dimitriades, 1981). Attention will therefore be paid here to the chemical transformation of anthropogenic hydrocarbon emissions.