Studies in Environmental Science 21

AIR POLLUTION BY NITROGEN OXIDES

Studies in Environmental Science 21

AIR POLLUTION BY NITROGEN OXIDES

Proceedings of the US–Dutch International Symposium, Maastricht, The Netherlands, May 24–23, 1982

Organized by the Ministry of Health and Environmental Protection in The Netherlands and the Environmental Protection Agency in the United States of America

edited by

T. Schneider

National Institute of Public Health, 3720 BA Bilthoven, The Netherlands

and

L. Grant

Environmental Protection Agency, Research Triangle Park, NC 27711, U.S.A.

ELSEVIER SCIENTIFIC PUBLISHING COMPANY
Amsterdam – Oxford – New York 1982

ELSEVIER SCIENTIFIC PUBLISHING COMPANY
Molenwerf 1
P.O. Box 211, 1000 AE Amsterdam, The Netherlands

Distributors for the United States and Canada:

ELSEVIER SCIENCE PUBLISHING COMPANY INC.
52, Vanderbilt Avenue
New York, NY 10017

Library of Congress Cataloging in Publication Data
Main entry under title:

Air pollution by nitrogen oxides.

(Studies in environmental science ; 21)
Includes index.
1. Air--Pollution--Congresses. 2. Nitrogen oxides--Congresses. I. Schneider, T. (Tonny), 1935-
II. Grant, Lester. III. Netherlands. Ministerie van Volksgezondheid en Millieuhygiëne. IV. United States. Environmental Protection Agency. V. Series.
TD885.5.N5A53 1982 628.5'32 82-16317
ISBN 0-444-42127-0

ISBN 0-444-42127-0 (Vol. 21)
ISBN 0-444-41696-X (Series)

Printed in The Netherlands

FOREWORD

The U.S.-Dutch International Symposium on Nitrogen Oxides, was held in Maastricht, The Netherlands, May 24-28, 1982, as one of many events commemorating the bicentennial of United States-Dutch friendship. The conference was attended by more than 300 people from 17 countries, including the United States of America, The Netherlands, other European nations, and Japan. Among the attendees were governmental administrative officials, research scientists, air pollution control experts, industrial representatives, members of environmental-conservation groups, news media representatives, and students from academic institutions of several European nations. Conference attendees heard more than 70 presentations, which included statements by U.S. and Dutch government officials made during an opening session attended by Her Majesty, Queen Beatrix, and subsequent technical papers reviewing state-of-the-art research and development information related to national and international policies on nitrogen oxides (NO_x) in the United States, The Netherlands, elsewhere in Europe, and Japan.

These proceedings contain texts of opening statements made by U.S. and Dutch government officials and the technical papers presented at the Symposium, as well as summaries of major exchanges of information that occurred during session discussion periods or as part of panel discussions. In general, the organization of the proceedings follows the format of the Symposium, which (in addition to the Opening Session) was divided into nine sessions that focused on the following topics: Overview of the NO_x Environmental Problem; Basics of NO_x Formation and Inventories of Sources; Monitoring Strategies and Exposure Assessment; Transport, Transformation and Removal of NO_x; Health Effects of NO_x; Ecological and Biological Effects; Stationary Source NO_x Control Technology; Mobile Source NO_x Control Technology; and Policy on NO_x Control and Implementation.

It would be very difficult to provide a comprehensive summary of the extensive and diverse array of information exchanged via the many excellent papers presented and discussed, both formally and informally, during the Symposium. However, certain highlights and impressions derived from Opening Session statements and associated discussions, as well as some major points of interest emerging from the later technical sessions, are worth noting here in order to convey an overall sense of the nature and scope of the information exchanged at the Symposium and contained in these published proceedings.

Especially noteworthy at the outset is the fact that considerable commonality of shared interests and views regarding environmental protection matters became evident among U.S., Dutch, and other conference participants, both as expressed in Opening Session statements and as reflected by further discussions during the course of the meeting. It was particularly encouraging to find agreement on several important concepts underlying approaches to environmental matters currently much discussed in the U.S., The Netherlands, and elsewhere. For example, the idea that environmental protection should be a shared responsibility among society as a whole, industry, and governmental entities appeared to be well accepted as a common view among the conference participants.

Also quite impressive was the commonality of views regarding the importance of adopting long-term perspectives in approaching environmental protection matters. More specifically, costs incurred now for environmental protection were stated to be wise investments for the future, with later returns to be gained not only in terms of health and ecological protection but also future economic benefits as well. It was noted that foresightful investment now in industrial planning and appropriate environmental controls should help to avoid potentially far higher later remedial clean-up costs or serious damage to human health or ecosystems. In that regard, attention was directed to statements made by Her Majesty, Queen Beatrix, in an earlier 1982 address before the Congress of the United States, in which she noted that "costs now are a condition for gains tomorrow" and "it is a tremendous challenge to promote economic prosperity and, simultaneously, to protect the environment."

Looking toward the future in meeting such a challenge, further agreement was apparent in terms of recognizing the need to develop improved approaches by which to harmonize environmental protection efforts with economic expansion or development and to assure that neither type of activity inordinately overshadows the other. That is, neither total emphasis on economic development without regard to its environmental impact nor emphasis on environmental protection without any regard to implementation costs and economic impact appear to be warranted. Rather, evaluation of alternative ways by which a balance between economic and environmental goals may be achieved in a cost-effective manner is necessary. Consistent with this, efforts are underway in the United States to seek increasingly more cooperative relationships between industry and government (more characteristic in the past of the European than the American scene) so that truely necessary measures for environmental protection can be more easily agreed upon and implemented in the most cost-effective manner. Thus, while specific measures may understandably differ from country

to country, because of different economic conditions and environmental problems, the basis for such actions derive from a shared common goal of harmonizing environmental protection and economic development activities.

Interest was expressed by both U.S. and Dutch representatives in exploring potential areas of future cooperation between the United States and the Netherlands regarding environmental protection matters. This includes a proposal to hold within the next few years another U.S.-Dutch Symposium (at a site in the United States), the topic of which would be of mutual interest and deal with another class of ubiquitous air pollutants (e.g., sulfur oxides or particulate matter) of concern in the United States, The Netherlands, and elsewhere. That international conference, it is viewed, would represent a joint U.S.-Dutch contribution to all nations in common efforts to address a specific environmental problem. As such, it would effectively build upon the foundation established by the present symposium on nitrogen oxides, which provides a model for close contact and collaboration between nations to foster information exchange on environmental protection matters of shared concern.

Turning to selected highlights emerging from the technical paper sessions of the present NO_x Symposium, the session on "The NO_x-Environmental Problem" included a series of general overview papers, which identified key issues of current interest and debate in regard to nitrogen oxides as an environmental problem and concisely noted examples of new research advances emerging in recent years or about to be published of importance in helping to resolve the identified issues. The overview papers drew attention to important issues now extant in regard to atmospheric processes and cycling of NO_x compounds in the ambient air, as well as ecological and health effects associated with exposures to nitrogen oxides and related compounds. Subsequent technical sessions included papers providing more detailed information and evaluations of specific problems and new research findings bearing on issues noted in the overview papers. In addition, other technical sessions focused on the implications of the newly emerging information for formulation of national and international NO_x regulatory policies and on advances in NO_x control technology.

In the session on "Basics of NO_x-Formation and Inventories of Sources," information was presented on chemical reactions involved in the formation of nitrogen monoxide (NO) in association with combustion of nitrogen-containing fuels such as coals and heavy fuel oils. Physical factors affecting the rate of chemical reactions leading to NO formation were also delineated, including temperature, pressure, species concentrations, and residence time. All of these are influenced by equipment design parameters (such as burner geometry,

combustion chamber size, and wall cooling mechanisms and rates) and by operational parameter (such as fuel preparation, fuel injection, mixing of fuel and air, and burner load). More specific information was provided on NO formation associated with fuel burning in motor vehicles, NO_x abatement strategies for mobile sources, and inventories of NO_x emissions from mobile and stationary sources in the United States and The Netherlands. The need for more detailed emission inventories to provide reliable inputs for the latest generation of long-range transport models for nitrogen oxide chemical cycles was also noted; and progress was reviewed in regard to development of such inventories and their use in joint U.S.-Canadian modeling activities concerning long-range NO_x transport.

In the session on "Transport, Transformation, and Removal of NO_x," information was presented on physical transport and chemical transformation of NO_x compounds in the ambient air. Important features of processes involved in the wet and dry deposition of NO_x substances, especially nitrate compounds, and interacting meteorological factors affecting the rates of deposition were also discussed. Of special interest were major advances reported in the development of improved mathematical models incorporating simulation parameters reflecting new knowledge regarding NO_x emissions, transport, and transformation processes and useful in predicting likely rates of NO_x concentrations at sites proximal or distal to to stationary or mobile emission sources. These include advances in: mathematical modeling of NO_x emissions and resulting NO_x concentrations on an urban scale as applied to selected areas in the United States and Europe; modeling of mesoscale transport of NO_x and resulting short- and long-term NO_x concentration levels in The Netherlands; and modeling of long-range NO_x transport phenomena constituting important determinants of NO_x concentrations and deposition in the United States and Scandinavia. Some case studies comparing modelled (predicted) and measured (observed) NO_x concentrations were described, which provided valuable information by which to evaluate the relative reliability and accuracy of several of the modelling approaches that were presented during the session.

NO_x monitoring systems in the United States, The Netherlands and Japan were described in the session on "Monitoring Strategies and Exposure Assessment." Several speakers noted the problem regarding intercomparability of monitoring results where monitoring data have to be used on a large scale. That is, use of different air monitoring techniques in different countries or sites within a country make it difficult to compare the respective NO_2 levels reported. Efforts toward harmonization of monitoring techniques among European countries are now underway both by a technical committee of the ISO and action

programmers of the European Community, as described in this session. In addition to discussion of outdoor monitoring information, important indoor NO_2 exposure information was also provided. Specifically, certain exposure assessment studies indicate that the actual exposure of people to NO_x is largely determined by the concentration of NO_x in the homes. Where gas appliances are used for heating, cooking, and other household applications, the exposure of residents to NO_2 and certain other indoor air pollutants is higher than for people living in homes using electric appliances, with indoor NO_2 levels in gas homes typically being higher than outdoor ambient NO_2 levels.

The session on "Health Effects of NO_x" included several reports on important new human clinical inhalation studies that extend our knowledge about dose-effect relationships for short-term NO_2 exposure effects on lung function in both normal and potentially sensitive (asthmatics and bronchitics) human subjects. The effects reported are typically acute in nature and rapidly reversible in terms of respiratory function. These studies do not allow us to evaluate the long-term, perhaps more permanent effects, of frequently repeated short-term exposures or chronic continuous exposures. Such effects are better evaluated, experimentally, by animal toxicology studies. Five of the eleven presentations in this session concerned effects of NO_2 on non-respiratory functions of the lung. These included studies which demonstrated increased susceptibility to infection by microorganisms due to the impact of NO_2 on lung defense mechanisms. This effect appears to be as much (or more) related to the pattern of exposure than just to total dose, with repeated short-term exposures to relatively high NO_2 concentrations (>1.5 ppm) being very effective in increasing susceptability to respiratory infection. Also, capillary cellular damage due to NO_2 was demonstrated, as measured by prostaglandin E_2 uptake, metabolism and release; and a variety of other biochemical effects as well as morphological changes in the lung were found after various NO_2 exposure regimes. These studies demonstrate potential acute effects as well as chronic implications of NO_2 toxicity. Approaches were also described by which advances are now being made in better determining crucial comparable dosimetry relationships between animal and human exposures to NO_2, and it is hoped that this will permit more credible extrapolation of animal study results to humans. In a later session concerning "Policy on NO_x Control and Implementation," summarized below, interesting comparisons were made between the interpretation of newly available health effects data by expert committees working to develop criteria for standard setting for NO_x in Japan, the U.S., The Netherlands, and elsewhere in Europe.

In the session on "Ecological and Biological Effects," new information on effects on plants of nitrogen oxides (either alone or in combination with SO_2 or O_3) was presented. Of particular interest were results concerning NO_2 effects on crop growth and productivity. In addition, new information was presented concerning the effects of acid rain on ecosystems in the United States, Scandinavia, and The Netherlands; and the potential contributions of NO_x emissions to such problems were noted. In another session on transport, transformation and fate of NO_x compounds, the role of NO_x emissions in acid deposition phenomena was discussed in more detail; and the long-range transport of transformation products (especially nitrates) to quite distant deposit sites, as well as local deposition, were noted. In the later session on regulatory approaches, the need for a multinational, European approach to control of NO_x and SO_2 emissions was emphasized in order to deal adequately with the transboundary long-range transport problem.

In the session on "Stationary Source NO_x Control Technology," two general types of stationary source NO_x control technology were discussed: combustion and post-combustion (or nitric acid tail gas) treatment processes. Much effort has been devoted during the past 15 years to development and commercialization of various control technologies. The combustion modification (CM) approach has emerged as a technology which can provide moderate to substantial NO_x reductions at relatively low cost. However, post-treatment or flue gas denitrification is now often employed in addition to CM where a higher level of control is required, even though higher capital and operating costs are incurred. Papers presented in this session described CM and post-treatment techniques in detail, with several novel concepts being delineated as well as more conventional techinques.

During the session on "Mobile Source Control Technology," information emerged on two issues of considerable interest and importance especially to The Netherlands and European community. Firstly, effective NO_x control does not appear to be possible based on current European regulations, because of the test cycle properties used therein. This cycle was developed 15 years ago mainly in view of problems with carbon monoxide (CO) in inner city areas. A proposal to help solve the problem of effective NO_x control, however, was made during the session. Secondly, it was also reported that there are a variety of technologies available to reduce NO_x to varying degrees (from significant to very drastic). Concerning the state-of-the-art of these technologies, some are only now in an experimental stage, while others seem far closer to being fully applicable in a few years and still others are now in full mass production. These technologies exert no negative impact on fuel consumption. In

some cases, even clear improvements are noted in fuel efficiency. Only in the case of very extreme reductions in NO_x can a negative effect occur. These technologies may of course, increase costs somewhat, especially when increased precision of machining operations is required in production processes. However, such increased costs will likely be offset by reduced fuel bills.

In the final technical session, concerning "Policy on NO_x Control and Implementation," information was presented regarding the existing United States National Ambient Air Quality Standards (NAAQS) for nitrogen oxides, strategies currently being employed for implementation of those standards, and the status of attainment of the standards. Also summarized was key information derived from reevaluation of criteria for health and welfare effects associated with ambient air exposures to NO_x compounds (as contained in the recently completed revised edition of the U.S. EPA document Air Quality Criteria for Nitrogen Oxides) and the implications of this latest criteria review for possible revision or retention of the existing U.S. NAAQS for NO_x. Analogously, the bases for NO_x standards and control policies in The Netherlands, elsewhere in Europe, and in Japan were described and discussed, and papers were presented on the views of some affected industry groups in regard to NO_x control policies and their implementation in the United States and The Netherlands. A wide-ranging panel discussion on NO_x Control Policy and Its Implementation then ensued, which included an open and highly interesting exchange of views among knowledgeable panelists drawn from federal and/or provincial (state) levels of government and affected industry groups in the United States and The Netherlands.

The excellence and value of the Symposium, as reflected by the high quality of new research findings presented and discussed in the technical sessions summarized above, was further enhanced by numerous other events and aspects associated with the meeting in Maastricht. Among these were included an excellent set of Technical Exhibits attached to the conference, at which several new technological advances in NO_x monitoring equipment and analytical approaches were displayed and explained along with books, research techniques and equipment, and other materials related to NO_x and other air pollutants. There were also numerous social events, including the Symposium Dinner and several receptions associated with the meeting, at which the very kind hospitality of our Dutch hosts was extended to the delight and appreciation of the conference attendees. In addition, several very informative optional excursions were arranged for interested participants to visit and learn about sites of historical, cultural and commercial interest in Maastricht, surrounding areas of Limburg Province, and elsewhere in The Netherlands and nearby areas

of Germany. Also of note was the issuance by Sphinx-Ceramique of a limited-edition set of ceramic tiles commemorating the U.S.-Dutch Bicentennial and the holding of the associated NO_x Symposium in Maastricht.

The successful conduct of an international conference of the magnitude of the U.S.-Dutch NO_x Symposium in Maastricht (which provides the basis for these published proceedings) represents an enormously complex and difficult undertaking, one which drew upon the talent, generosity, and dedication of numerous individuals and groups to whom the present Editors and Co-Chairmen of the Symposium are deeply indebted. It would be impossible to acknowledge here all of the many people and organizations who contributed in many diverse ways to the organization and conduct of the Symposium and associated events, as well as to the subsequent preparation for publication of the present proceedings. We do wish, however, to express our sincere and deep appreciation to at least some specific individuals whose very hard work or gracious hospitality contributed in an exceptional manner to the success of the Symposium and the comfort of the participants.

We are highly appreciative of the interest and involvement in the Symposium of Her Majesty, Queen Beatrix, and the participation of Mrs. J. J. Lambers-Hacquebard, State Secretary of The Netherlands Ministry of Public Health and Environmental Protection. We are equally appreciative of the participation of Mrs. Kathleen Bennett, Assistant Administrator, Office of Air, Noise and Radiation, the U.S. Environmental Protection Agency, who attended the Symposium as the representative to the meeting of President Reagan and Ms. Anne Gorsuch, Administrator of the U.S. Environmental Protection Agency. We are also appreciative of the participation of Mr. W. D. Boggs, Chargé d'Affaires, Embassy of the United States of America in The Netherlands, on behalf of the United States Ambassador to The Netherlands.

The considerable generosity and assistance of several other Dutch officials at various levels of government are worthy of note here as providing significant contributions to the success of the Symposium. These include: Dr. J. Kremers, Commissioner of the Queen for the Province of Limburg; Mr. M.H.C. Lodewijks, the Commissioner's Representative, and Mr. Alphonse Brüll of the Commissioner's Staff; Mr. A.I.M.H. Baeten, Burgomaster of Maastricht, and his associate Alderman Peeters. The invaluable assistance of Mr. David Strother and Mr. Fred Zandvliet, of the Offices of International Affairs of the U.S. Environmental Protection Agency and The Netherlands Ministry of Public Health and Environmental Protection, respectively, are greatly appreciated as well.

Organization of the U.S.-Dutch NO_x Symposium and coordination of preparation of the present proceedings would have been impossible without the aid of the members of the Symposium Advisory and Organizing Committees listed later in the present volume; and we are deeply indebted for the excellent contributions of all members of those committees. We are especially appreciative of the very extensive and dedicated work of Dr. J. van Ham, of SCMO-TNO, who served as Secretary for the Symposium, and of Dr. Si Duk Lee, Associate Director of the U.S. EPA Environmental Criteria and Assessment Office, who served as Symposium Coordinator within the U.S. EPA. The valuable technical inputs of Drs. L. C. van Beckhoven and S. Zwerver, of the Organizing Committee, are also greatly appreciated, as is the excellent organization and conduct of the Ladies Programme by Mrs. M. Schneider. Of extremely great value also are the efforts of the members of the Symposium Bureau, who assisted in registration of Symposium attendees and preparation of the present proceedings for publication. These include: Mrs. P.W.A.M. Venis-Pols; Mrs. O.M.B. van Steenis; Mrs. M.C.M.A. Poelen-Jansen; and Mrs. C.H.M. Olij.

We look forward to future cooperative efforts involving many of the above individuals and organizations and hope that the high standard of quality and excellence established by them in relation to the present project can be matched by such future endeavors.

Dr. Lester D. Grant,
U.S. Environmental Protection Agency,
Research Triangle Park, NC (U.S.A.)

Dr. Toni Schneider,
Ministry of Public Health
and Environmental Protection, RIV,
Bilthoven (The Netherlands)