


**AIR QUALITY
RESEARCH BRANCH**

ANNUAL REPORT

1996-97

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Compiled by
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June 1997

Air Quality Research Branch
Atmospheric Environment Service
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AIR QUALITY RESEARCH BRANCH

ANNUAL REPORT

1996/97

1.0 FOREWORD

1.1 AIR QUALITY RESEARCH BRANCH

- D.C. McKay

The Branch has been involved in a full and varied research program looking at air quality issues. Even though the report details results in the individual issue areas, these atmospheric issues are inter-related with each other, as well as being inter-related with the other portions of the environment. Therefore, projects are undertaken in partnership with colleagues from other organizations and from different scientific disciplines not only in Canada but worldwide.

This year the Atmospheric Sector report of the 1996 Canadian Acidifying Emissions Assessment was completed. This report indicated the direction that the atmospheric research program had taken in the last few years based on recommendations of an 1990 assessment report. Questions being addressed are: what is the impact to date of emission control programs; what is the projected impact of these controls; and are existing limitation measure adequate to protect human health, forestry and visibility.

Particulate matter effects human health and visibility. Based on an international agreement with the United States, the Branch has conducted investigations into visibility degradation due to particulate matter. These investigations have focused on historical visibility and on the contribution of aerosols to reductions in visibility. Human health studies have shown that aerosols have an influence on the health of humans through the relationship of hospital admissions to the level of aerosols in the atmosphere. To understand more fully the effect of aerosols on the environment, the Branch is moving into particulate matter research through investigations of particle size effects and of their relationship to light scattering and to deposition.

Chemical and physical atmospheric observations are important in defining the temporal and spatial trends in atmospheric conditions. The Branch has many sites throughout Canada where measurements of atmospheric constituents are being made. These measurements are augmented by data from other agencies and from intensive short-term field studies. Even though the Branch focuses on several air quality issues, all issues are inter-related and so are the data. Efforts are being made

to consolidate the monitoring network so as to gain the most data from the resources available.

The measurement campaign to monitor and study the chemistry and dynamics of the arctic stratospheric ozone layer in spring 1997 showed that the polar vortex was late in establishing itself. However, once it did, it remained strong, cold and centered over the north pole much later into spring than in previous years. From the frequent observations of polar stratospheric clouds, it is inferred that chemical and dynamic effects played a major role in decreasing the ozone to the very low values measured.

1.2 CARE

- F.A. Froude

CARE (Centre for Atmospheric Research Experiments) is the Branch's window on the atmosphere. Situated in rural southern Ontario, it hosts a combination of atmospheric measurement programs with national and international organizations by supporting the preparation and collection of field samples for environmental studies. More information on the various measurement programs are available on the CARE Home Page through the Branch's web site.

At present there are over 40 research projects operating at CARE which range from the measurement of precipitation using the AES standard collectors, the collection of ambient concentration of toxic chemicals in air, to the development of new state-of-art techniques for the mapping of air pollution episodes using LIDAR (a combination of laser light source coupled with a telescope to measure reflected backscatter energy).

CARE an EMAN site with 3 bio-diversity plots

Recently, CARE was initiated as an EMAN (Ecological Monitoring and Assessment Network) site with the development of three bio-diversity SIMAB (Smithsonian Institution/UNESCO-Man and the Biosphere) plots: two plots (a mixed hardwood and a red pine) at CARE and a plot at the nearby Tiffin Centre for Conservation. Tiffin, in cooperation with CARE, has developed an environmental educational program which is used by area schools and community groups.

Additions to the air quality measurements include: a condensation particle counter for measuring the total number of particles; a tapered-element oscillating measurement system for measuring total suspended particles; an ambient carbon particle monitor; and vapour phase mercury measurements at Egbert and Point Petre.

1.3 PROGRAM REPORT

- K. Ford, L. Grittani and M. Still

On April 1, 1996, the Branch had 109 employees. With employees leaving due to workforce adjustment and with the hiring of new skills, the Branch ended the year with 108 employees. During the year, 17 visiting scientists from all parts of the world collaborated with the Branch at its facilities. Of these 11 were on long-term assignments. To assist the Branch in its research activities, 38 students were hired under various student programs.

Of the 119 trips that Branch staff took, 62 were to international conferences, symposiums, workshops and business meetings. The remaining trips were within North America.

The Branch began the year with an allocation of nearly \$14,600K, which was comprised of \$6,000K for salaries, \$7,500K for operational costs and \$1,100 for capital purchases. Supplementing this budget, the Branch received about \$2,700K from numerous other parties to aid research activities. Some of the larger amounts were from the DOE Strategy Capital Fund for four strategy capital projects (\$1,370K), from the DOE Ontario Region for the Great Lakes Action Plan (\$690K), from DIAND for the Northern Contaminants program (\$270K) and the from the Climate Research Branch for the Branch's climate program (\$210K).

Phase II of laboratory implementation commences

This year, the Branch initiated the second phase of its plan which will include bringing the remaining off-site laboratories at York University to the main laboratory building in North York. Public Works & General Services Canada (PWGSC) hired the architects, Shore Tilbe Irwin & Partners (STI&P), to commence work in August. STI&P held initial meetings with the laboratory managers to discuss the general concepts of the required work and to detail the information that they wished from the managers. This was followed by in-depth discussions with each manager, as well as in-lab visits to gain knowledge on the operations and equipment used. By the end of March 1997, the laboratory requirements had been finalized. As well as the plans for the laboratory space,

changes were proposed to the original phase to enhance the operation of the building. The design included the addition of an extra link on the 1st floor between each building and an external loading/unloading area for small vehicles. Occupancy is planned for early 1998.

1.4 AWARDS

- M. Still

3 employees receive All-Seasons Research Award

Since 1988, the Branch has been presenting the All-Seasons Research Award to staff who have forwarded the goals of the Air Quality Research Program. To date, 31 such awards have been presented, including staff from other sections of Environment Canada. In 1966, three employees were recognized for their work.

- Sadiq Ahmed who developed the ozone module in the Research Data Management and Quality Control System (RDMQ). He was relentless in ensuring that every detail of the protocols, which he developed over the last few years, would be accurately applied. The module of the RDMQ software package now reflects this attention to detail that was so inherent in Sadiq's work for the Branch. Sadiq's contribution was well outside his expected duties and this contribution has ensured the continuing high quality of atmospheric data.
- Tom Dann who was involved in the NO_x/VOC Science Program. He has led the Science Program's Ambient Data Analysis Work Group and, through his co-ordination and his contributions, has edited the final report. This has been a major accomplishment for the AES-led NO_x/VOC Assessment. Tom has also provided willing support by providing on-request ambient data analyses for ground-level ozone, nitrogen oxides, volatile organic compounds and particulate matter. These analyses have allowed Branch and AES staff to make effective scientific presentations to Environment Canada management, to other government departments and to stakeholders.
- Harold Neumann who established the Borden Tower site. This site has been used for several ground-breaking studies on turbulent exchange of energy and trace gases. Data collected at this site has enabled modellers to improve the dry deposition parameterization in the Acid Deposition & Oxidant Model (ADOM). Harold co-led the AES team at the BOREAS study which has produced important information on energy and carbon budgets of boreal forests. Harold is an internationally-respected expert

on boundary layer processes, air pollution and micrometeorology. For many years he has been on the editorial board of Agricultural & Forestry Meteorology.

1.5 BRANCH RESEARCH ACTIVITIES

1.5.1 Flow Over Complex Terrain - J.L. Walmsley

Preliminary work on improvements to the *Guidelines for Estimating Wind Speed Variations in Complex Terrain* was undertaken using PERD (Panel on Energy Research & Development) funding under a Collaborative Research Agreement with Prof. Peter Taylor of York University. A new form of the upstream profile has been coded. Work is progressing on incorporating the effects of stable stratification.

1.5.2 Wind Energy - J.L. Walmsley

In collaboration with Dr. J.R. Salmon of Zephyr North, a paper describing tests and applications of the *Wind Correlation Model* of Walmsley & Bagg (1978) was prepared for presentation and publication. The associated software provides a capability to relate short-term wind speed and direction measurements at a candidate wind-turbine site to long-term wind climatology at a nearby weather or climatological station. The method should have wider application to the general case of two-site correlation of winds.

2.0 THE ISSUES

2.0.1 ACIDIFYING EMISSIONS - S. Venkatesh

The overall objective of the Program is to study the transport, transformation and fate of acidifying emissions with the intent of providing advice and guidance for reducing risks to human health and the environment.

This year, the research program addressed three areas:

- the examination of the linkages between acidifying emissions and the resulting air concentrations and deposition of acidity, and between sulphur and nitrogen species in order to determine whether existing control measures are being effective in reducing the environmental stress;
- the investigation of areas of uncertainty identified by the Science Assessment (The Science Assessment is part of an effort by the Federal/Provincial National Air Issues Coordinating Committee to derive a strategy for the

management of acidifying emissions in the post-2000 era) which preclude definitive statements on certain questions, and the analysis and interpretation of additional policy questions raised by the assessment; and

- the exploration of the linkages between certain aspects of this research program and research on the causes of other environmental issues, e.g., linkages between acidifying emissions, and the formation and loss of particles.

[For description of present projects, see Section 2.1]

2.0.2 HAZARDOUS AIR POLLUTANTS - K. Puckett

The research goal is to understand the environmental cycling of air toxics with a view to better predict the behaviour of these chemicals. This research includes considerations of emissions, transport, transformation and deposition of these chemicals. These air toxics include pesticides, industrial chemicals, such as polychlorinated biphenyls (PCBs), and products of combustion, e.g., polycyclic aromatic hydrocarbons (PAHs). There are three main thrusts to this Program, namely, support to Annex 15 of the Great Lakes Water Quality Agreement (GLWQA), the DIAND-sponsored Northern Contaminants Program, and Keeping Toxics out of the Environment Program.

For the GLWQA, there are two thrusts, a surveillance/monitoring component and a process research component. Surveillance is accomplished through the Integrated Atmospheric Deposition Network (IADN) which has been in operation since 1988 and consists of two master stations (Point Petre and Burnt Island) and a satellite station (Egbert). Data are analyzed from this network to provide estimates of loadings to the Great Lakes, and to assess the sources and pathways of toxic chemicals.

The goal of the Northern Contaminants Program is to reduce, and wherever possible to eliminate, contaminants in traditionally harvested foods, while providing information that assists informed decision-making by individuals and communities in their food use. The Branch assists in this context by identifying and quantifying some of the contaminants, and determining the pathways and processes that deliver these contaminants to the north. One activity has been the measurement of CHBs (chlorobornanes) and HCHs (hexachlorocyclohexanes) in arctic air and water. These data are used to investigate the air-sea gas exchange rates of CHBs and HCHs. Other initiatives are investigating the physico-chemical properties of CHBs and the changes of OCs (organochlorines) in the air-water-food chain.

Both the GLWQA and the Northern Contaminants Programs have common activities. Both involve research to improve the understanding of the processes involved, to

determine the fate of these compounds, and to develop models of transport and fate. Specific activities are:

- investigating mesoscale and global models for their applicability for toxic chemical transport;
- collecting usage inventory of priority chemicals;
- developing re-emission models (volatilization) of chemicals from soils;
- improving knowledge of air-water exchange and dry deposition processes; and
- improving understanding of photochemical processes.

The component entitled "Keeping Toxics out of the Environment" is promoting the development of a national approach to hazardous air pollutants (HAPs). Besides the continuing work on the monitoring of mercury, this effort will define the scope of the HAPs problem, and will indicate the temporal and spatial scales involved for various compounds.

[For description of present projects, see Section 2.2]

2.0.3 TROPOSPHERIC OZONE

- M. Lusic

The goal of the NO_x/VOC (Nitrogen oxides/Volatile organic compounds) Science Program is to characterize photochemical oxidants in Canada, and the underlying physical and chemical processes that control their occurrence, such that any emission control policy measures can be based on sound scientific information.

This year, the major focus was on the following two areas:

- Scientific Assessment of Tropospheric Ozone in Canada: Much effort was devoted to scientific analyses and writing Work Group reports for this Assessment. Some of the results have already been used in policy development and in planning the next steps in the NO_x/VOC Management Plan.
- Eastern Canadian Oxidants Field Study (NARSTO-CE): In coordination with the NARSTO-Northeast (North American Research Strategy for Tropospheric Ozone-Northeast) campaign in the Ozone Transport Region of the United States and two regional campaigns in eastern Canada (the Southern Ontario Oxidants Study, SONTOS, and the Montreal Area Oxidants Study, MAOS), surface and upper air measurements were carried out during June and July 1996 in the Windsor-Quebec Corridor and Southern Atlantic Region, with the primary objective of providing data for further evaluation and improvement of the long-range transport oxidant models. Two questions were of particular interest: how accurately do the regional oxidant models developed under the NO_x/VOC Management Plan's Science Program represent the atmospheric chemistry

of oxidant formation in an eastern North America setting?; and how well does the MC2 meteorological driver in these oxidant models reproduce the dynamics of the nocturnal and the marine boundary layer? Preliminary results from NARSTO-CE, and further data analysis and model evaluation activities were discussed in a series of workshops in December 1996.

In addition, support continued for a number of ongoing projects to assess the effectiveness of emission controls through air monitoring of ozone and nitrogen compounds at regionally-representative CAPMoN (Canadian Air and Precipitation Monitoring Network) sites and Mt. Sutton, and data analyses using NAtChem (National Atmospheric Chemistry) and Oxidants databases. In the area of model improvement and evaluation, there were several activities, including further model performance evaluation in the Lower Fraser Valley region and in eastern Canada, and work on a meteorological aggregation scheme for conducting emission scenario runs so that results can be based on a variety of meteorological conditions rather than just one episode.

[For description of present projects, see Section 2.3]

2.0.4 STRATOSPHERIC OZONE

- D. Wardle

In line with the goal to reduce uncertainties in predictions of climate change and to help Canadians adapt to current and expected changes, the focus has been the interaction of solar electromagnetic radiation with the atmosphere. This focus relates both to the shielding of the earth's surface from harmful ultraviolet radiation and to the supplying of energy from the sun to the earth's surface. Specific tasks have addressed the stratospheric chemistry which effects ozone, the quantification of long-term changes in stratospheric ozone and in radiation properties, and the determination to what extent these changes are anthropogenic and controllable.

All locations outside the tropics have experienced significant losses of column ozone during the last 30 years due to the introduction of chlorofluorocarbons (CFCs) and other ozone-depleting substances. Tropospheric CFC concentrations are no longer increasing but recovery in the stratosphere will be delayed due to the time required for migration of CFCs from the troposphere to the mid-stratosphere where they are converted into inorganic chlorine molecules by ultraviolet radiation.

The main activities have been centred on:

- the Bratt's Lake radiation observatory;
- the National Atmospheric Radiation Centre;
- the World Ozone and Ultraviolet Radiation Centre;

- the Brewer ozone and ultraviolet radiation observation network;
- the Eureka observatory;
- aircraft campaigns; and
- satellite studies.

[For description of present projects, see Section 2.4]

2.0.5 CLIMATE CHANGE

- M. Lulis

The overall objective is to contribute to our understanding of greenhouse gases and aerosols - their trends, budgets and role in climate change - by carrying out measurements, modelling and process studies with a Canadian focus, and by interfacing with other major international programs.

The major objectives of the measurement component are to obtain background concentration levels of greenhouse gases, to evaluate the global transport and transformation of climatically active trace species by identifying their source regions and major sinks, to determine the impact of emission control strategies and to support the WMO Global Atmosphere Watch Program.

Resources were directed toward the ongoing measurement activities at Alert, Fraserdale and the two coastal sites (Sable Island and Estevan Point). At these four sites, some or all of the following measurements are made by AES staff and other collaborating scientists: CO₂, CH₄, N₂O, O₃, PAN, CO, CFCs, radon, C and O stable isotopes in CO₂ and CH₄, aerosol scattering and chemistry, PAHs, hydrocarbons, mercury and meteorological parameters.

In addition to the above activities, other projects were supported:

- Canadian support was provided for the cooperative project to implement the Chinese GAW station at Waliguan Mountain. This station will provide, for the first time, background levels of aerosol chemistry and other atmospheric constituents from a remote Asian continental site.
- Analytical methodology and validation techniques were developed for the analysis of C isotopes from routine flask samples of CO₂ in air. Methodologies were also developed for analysis of C and O isotopes in atmospheric CO, and applied to a regional-scale source apportionment study. A feasibility study was completed on analysing C isotopes in the carbonaceous components of atmospheric aerosols.
- Field studies were undertaken into the fluxes of DMS (dimethylsulphide) and its oxidation products (SO₂, sulphates and methanesulfonate). Extended analytical capabilities permitted the measurement of other sulphur compounds, such as carbon disulphide, carbonyl sulphide and dimethyl disulphide.

- As part of a proposed international network of tower CO₂ flux sites, measurements of the exchange of CO₂ between the atmosphere and two different forest ecosystems in distinct climatic zones were initiated to determine the magnitude, and the seasonal and inter-annual variability of these fluxes which will be vital in providing constraints to models of the global carbon cycle, and in pinpointing the missing carbon sink.
- In addition, a Climate Research Workshop was held in December 1996 to answer the question on how best can the AES focus its atmospheric measurements research to answer priority issues in carbon cycle research (report available from M. Lulis).

There were several other climate-related modelling projects:

- Work continued on assessing the influence of anthropogenic sulphate and black carbon aerosols on northern hemispheric climate (and the Canadian climate in particular) by developing a high resolution northern aerosol regional climate model (NARCM) with anthropogenic aerosols as active constituents.
- Through PERD (Panel on Energy Research & Development), work continued on the development of 3D climate/carbon cycle coupled model. This work is being carried out in collaboration with Forestry Canada, Agriculture Canada, McGill University, the University of Toronto, and Japanese research institutes. As well, work on the validation and improvement of the 2D climate/photochemical coupled model continued, including dynamical/radiative/photochemical processes from the surface to 60 km. Collaborative modelling projects with Tohoku University in Japan included development of a multi-box global model to investigate cycling of CO₂ isotopes, estimation of latitudinal distribution of CO₂ and CH₄ sources and sinks, interpretation of atmospheric CH₄ measurements, and interpretation of CO₂, CH₄ and O₃ measurements taken by the Japanese in the Antarctic and Arctic.

[For description of present projects, see Section 2.5]

2.1 ACIDIFYING EMISSIONS

2.1.1 Aerosol Optical Properties and Visibility

- R.M. Hoff and L. Guise-Bagley

The objective was to determine the effectiveness of atmospheric aerosols in reducing visibility and to determine particle size effects and their relationship to light scattering and deposition.

The Branch operated four stations for assessing visibility reduction (Vancouver, Waterton Lakes, Egbert, and St.

Andrews, NB). One more year of nephelometer results will be obtained at an Alberta, an Ontario and a New Brunswick site with a view to relocation of two of the sites in 1998. This will aid in determining the aerosol characteristics (size, speciation and growth) which impact on the atmospheric visibility issue.

Study of optical properties doesn't end at esthetics but also impacts on air quality and human health

This year completed the third year of operation and preliminary results have shown that outside of urban areas, sulphate is a major component of the ambient aerosol, followed by organic aerosol and black carbon. The organic aerosol in remote areas, like St. Andrews, shows an annual cycle peaking with biological activity in the summer. In Vancouver, however, no cycle is seen and is probably due to the large portion of organic aerosols from anthropogenic combustion activities. Currently, projects are being investigated to speciate this organic aerosol. Visibility measurements at these sites are consistent with the historical visibility data. Computer models have been used to predict what future visibility may be if sulphur controls are more fully implemented.

Visibility degradation from aerosols is one of the most apparent forms of pollution to the public

This project has been achieved through collaboration with J.L. Campbell and Z. Nejedly (U. Guelph), K. McDonald (DOE P&N Region), S. Pryor (U. Indiana), R. Barthelmie (Riso National Laboratory, Denmark), D. Steyn (U. British Columbia), R. Hughes (New Brunswick Environment), and Y. Golestani and W. Malm (CIRA, Colorado State University, Fort Collins).

2.1.2 CAPMoN

- D. MacTavish, N. Lance, R. Braga, B. Kessler, B. Kiely, W. Kobelka and D. Ord

The Canadian Air and Precipitation Monitoring Network (CAPMoN) operated directly or in partnership 11 air filter measurement sites and 21 precipitation chemistry sites between April 1, 1996, and March 31, 1997. December 31, 1996, marked the closure of the Montmorency, Quebec, monitoring site. A regular calibration/inspector program (quarterly) was maintained at all sites with the help of the DOE regional staff, while network integrity was ensured by comprehensively auditing operations at 11 sites. A CAPMoN technical workshop was held in Niagara Falls in September 1996.

Participants from federal and provincial governments, as well as the Norwegian Institute for Air Research, ensured its success.

Partnerships continue with Nova Scotia Environment and the North West Territories Government in the operation of two precipitation chemistry monitoring sites. A new partnership between Manitoba Environment and the Branch has been established to ensure the continued monitoring of precipitation at Island Lake, Manitoba. Temporary cost-sharing agreements were reached with Pacific and Yukon, Prairie and Northern, Ontario, and Atlantic DOE Regions allowing for the continuation of 4 precipitation monitoring sites.

Over 22,000 samples analysed during the year

Chemical analyses and the compiling of CAPMoN laboratory data sets for the 1996 network samples were completed. This included the analyses of over 15,000 filters and 7,500 precipitation samples, involving more than 154,000 chemical determinations. Laboratory credibility was maintained by continued successful participation in international intercomparison studies including Environment Canada's NWRI/NLET Intercomparison, the WMO-GAW's Laboratory Intercomparison, the United States Geological Survey Intercomparison, and the Norwegian Institute for Air Research's European Measurement and Evaluation Program Laboratory Round Robin Study.

2.1.3 Quality Assurance and Data Management

- R. Vet, S. Iqbal, M. Shaw and W. Sukloff

As well as this program area, this project supports the NO_x/VOC (Nitrogen Oxides/Volatile Organic Compounds) program area. The CAPMoN (Canadian Air and Precipitation Monitoring Network) Quality Assurance and Data Management Program continued in support of the network. Performance audits were carried out at CAPMoN filter pack and ozone monitoring sites, and a formal audit report was produced. A first draft of a CAPMoN Quality Assurance Report was also produced. Finalized CAPMoN data were transferred to the National Atmospheric Chemistry Data Base, and quality assurance and data management support was provided NARSTO-CE (North American Research Strategy for Tropospheric Ozone-Canada East) field study.

2.1.4 Global Climatological Dispersion Model

- C.S. Matthias

The Global Climatological Dispersion Model (GCDM) is a time-dependent dispersion model in spherical

coordinates. In numerical form, it has finite differences in the longitude and latitude directions, and two layers in the vertical direction. It can be applied as a regional model to a portion of the globe, i.e., North America, or to the entire globe. The input meteorological data are climatological and represent mean conditions over a month, season or year. Data are available on a global 1 degree latitude/ longitude grid. The two layers of the model are the planetary boundary layer and the troposphere layer with respective thicknesses of about 1 km and 6.5 km.

Initial studies were carried out with a single layer model. It was first applied to examine sulphur concentrations and deposition over North America. Results are similar to measurements, except that the modelled 10 kg/ha/yr line for sulphate deposition in Canada is about 500 km south of the measured line. The GCDM was also applied globally in a qualitative study to test the numerical scheme as well as to calculate the sulphur distribution. Results were compared to 3D Lagrangian model results with both models using the same source inventory. Concentrations over source regions had similar values but the GCDM showed lower in the polar regions, and much lower in parts of the Atlantic and Pacific oceans. A 2-layer version of GCDM was used to calculate the global accumulation and distribution of krypton-85 over a 40-year period (a byproduct of nuclear fuel reprocessing). Results compared well to measured data and to a 3D Eulerian model output.

Model is simple in structure and concentrates on first-order physics and chemistry

The model is simple in structure and concentrates on first-order physics and chemistry. It has relatively modest input data requirements and runs quickly on a personal computer. It is applicable to a variety of investigations, such as continental to global dispersion for time periods of months to decades, the impact of cross-boundary transport for a variety of source scenarios, and the pesticide storage and movement within and between media.

2.1.5 Fogwater Deposition to High-Elevation Forests

- J.L. Walmsley

A method for estimating fogwater deposition to forests in mountainous terrain was developed using measured fogwater volume as input. This is a step towards the longer-term goal of a method that uses only routinely available meteorological data. The importance of this work is that, by using the MS-Micro wind flow model and an assumption about variation of liquid water content

(LWC) with height above cloudbase, spatial variations of fogwater deposition in mountainous terrain are possible. Calculated deposition rates were found to be typical of measured values in the scientific literature.

Estimation of LWC at one elevation in mountainous terrain using only routinely available data

The LWC available for deposition on the high-elevation forests of Roundtop Mountain was calculated by only using routine observations from the weather station at Sherbrooke, Quebec (78 km from Roundtop). The method uses the Classification and Regression Tree (CART) software and will later use the Neuro-Fuzzy Inference System (NFIS). Sample calculations with data from 1988-91 have now been done and more extensive tests are in progress. The importance of this project, if successful, will be the estimation of LWC at one elevation in mountainous terrain using only routinely available data. When combined with the methodology of Walmsley et al. (1996), estimates of the spatial variations in fogwater deposition should be possible from routine data.

This project was undertaken in collaboration with N. Urquizo, W.R. Burrows, R.S. Schemenauer and J.R. Brook.

2.1.6 Spatial Variability of Dry Deposition of Gases

- J.L. Walmsley

Sensitivity testing of the *MSFD-CONC* model code began in 1996/97. Preliminary tests uncovered a coding error which was corrected. The long-term goal is the estimation of dry deposition of up to four pollutants in complex terrain (hills and valleys, changes in surface cover and roughness) with the possibility of linear interactions among the pollutants. Until sensitivity tests have been completed, only one pollutant will be included.

2.1.7 Organic Atmospheric Sulphur

- S. Sharma and L.A. Barrie

The focus is to understand the role of the biogenic sulphur, mainly dimethyl sulphide (DMS), influencing concentrations of atmospheric aerosols. Marine and freshwater sources of DMS contribute significantly to the background atmospheric sulphate aerosol upon which anthropogenic aerosols are superimposed. Since 1993, several studies have been carried out in the oceanic and freshwater environments in order to assess the relative contribution of biogenic sulphur to the atmosphere.

In 1993, a collaborative study was done in the Gulf of St. Lawrence with Dr. M. Levasseur (Fisheries & Oceans)

and Dr. M. Gosselin (University of Quebec in Rimouski) to assess the contribution of DMS from the Gulf to the atmosphere. On an average, the Gulf of St. Lawrence contributed $4.6 \mu\text{mole}\cdot\text{m}^{-2}\cdot\text{d}^{-1}$ of biogenic sulphur (DMS) which at most is 16% of the anthropogenic emissions of sulphur contributed by the surrounding provinces.

An Arctic Ocean section expedition, lead by Canadian and Americans on two ice-breakers, Louis St. Laurent and the Polar Sea, that crossed the North Pole, took place in 1994. Fluxes of DMS from the Arctic Ocean were determined in collaboration with Drs. M. Levasseur, M. Gosselin (Rimouski) and T. Bates (NOAA, Washington). In the sub-Arctic ($50\text{-}70^\circ\text{N}$), the mean DMS was $11.3\pm 8.0 \mu\text{mole}\cdot\text{m}^{-2}\cdot\text{d}^{-1}$, whereas the central Arctic Ocean contributed an average to the atmosphere of $0.041\pm 0.051 \mu\text{mole}\cdot\text{m}^{-2}\cdot\text{d}^{-1}$ of DMS. A 2-dimensional, multilevel, photochemical model (Plummer et al., 1996) was used to estimate the atmospheric DMS concentrations. The model predictions of the atmospheric DMS were fairly comparable to the measured values.

Lakes contribute annual emissions of 1.2×10^6 kg S (DMS) to the atmosphere during the ice-free season

A two-year study was conducted in 1995 and 1996 to determine the contribution of DMS from the lakes of the Canadian Boreal Shield. A site in the Experimental Lakes Area, northern Ontario, was chosen to determine fluxes of DMS from the lakes during the ice-free season. This work was done in collaboration with Dr. Carol Kelly (U. Manitoba) and Dr. John Rudd (Fresh Water Institute, Winnipeg). The lakes of the Canadian Boreal Shield contribute annual emissions of 1.2×10^6 kg S (DMS) to the atmosphere during the ice-free season of 9 months. These emissions are 65% of the total sulphur released by land (vegetation, soil, forests).

2.2 HAZARDOUS AIR POLLUTANTS

2.2.1 IADN Data Analysis and Interpretation

- R.M. Hoff, K.A. Brice, F. Froude, P. Blanchard, J. Woods, C. Audette, B. Sukloff, B. Martin and J.F. Hopper

The focus has been to interpret the data from the IADN (Integrated Atmospheric Deposition Network) program, to provide estimates of loadings of toxics to the Great Lakes, to assess the sources and pathways of toxic chemicals, and to provide a quality-assured data set of the IADN results.

IADN has been in operation in the Great Lakes region since 1988. AES is responsible for the operation of two Master Stations (Point Petre and Burnt Island, Ontario) and one satellite station (Egbert, Ontario). Trace organics in air and trace metals in air are monitored every 12 days. In addition, meteorological conditions are determined at the site and trajectory modelling support is provided to determine potential sources of chemicals. Every two years, the Branch contributes to estimates of loadings of hazardous air pollutants (HAPs) to the Great Lakes. The IADN data is also used to determine trends in air concentrations and progress towards virtual elimination of toxics from the Great Lakes.

The Branch led the last published loadings review of the Great Lakes basin (Hoff et al., 1996) -- a publication which has gained widespread dissemination. This year, data will be contributed to the 1994 data year loadings review being conducted by the U.S. EPA and their contractor, the University of Indiana. Principle component analysis, conditional probability source function analysis and back trajectories have been examined to determine sources and source types affecting the Great Lakes basin.

Collaborators have been A. Bandemehr (U.S. EPA), R. Hites, B. Hillery, D. Cortes and D. Wagrowski (U. Indiana), C. Sweet (Illinois State Water Survey), G. Keeler (U. Michigan), W.M.J. Strachan and D. Burniston (NWRI), M. Neilson and C.H. Chan (EHD-OR), D. Orr (Ontario Ministry of Environment & Energy) and T. Dann (EPS).

2.2.2 Organics Analysis Laboratory

- K.A. Brice, N. Alexandrou, K. Su, R. Park and M. Shoeib

The Organics Analysis Laboratory (OAL) provides an in-house service for the determination of trace organic chemicals in air samples for AES responsibilities within the bi-national Integrated Atmospheric Deposition Network (IADN) of Annex 15 of the Great Lakes Water Quality Agreement. Target organic species include polychlorinated biphenyls congeners (PCBs), organochlorine pesticides (OCs) and polycyclic aromatic hydrocarbons (PAHs). Samples are regularly collected at three Canadian IADN sites: Point Petre, Burnt Island and Egbert.

Ambient samples for trace organics are collected using the PS-1 sampler, employing a glass-fibre filter (GFF) for particle retention, followed by a polyurethane-foam

(PUF) cartridge as a vapour-phase adsorbent. The volume of air typically sampled is approximately 350 m³, taken over a 24-hour period. The two sample components are analysed separately to provide "operationally-defined" information on phase distributions.

Because of the sample backlog, a contract was issued to DFO Winnipeg for assistance in the analysis of samples from Egbert. OAL has been closely involved with this contract by providing extraction protocols, standards for recovery evaluations and parallel analysis of selected "split" sample extracts from this set. Results from the DFO analysis are still being assessed, but numerous problems have been identified with anomalous recovery values for spiked samples and poor detectability.

Point Petre and Burnt Island data allow examination of trends and seasonal variability

The final product of the laboratory chemical determination procedures is a database containing the amounts of analyte detected for each sample, together with information on detection limits, lower calibration limits and preliminary qualifier flags. Using the AES Organics RDMQ (Research Data Management and Quality Control System), the "back-filling" of non-detects can be implemented, blank levels can be assessed, corrections for blanks can be performed, statistical information can be calculated for a variety of time periods, and correlations between atmospheric species can be investigated. Field sampling data are combined with the analytical laboratory results to compute actual ambient air concentrations.

OAL has also been very active in following its research mandate to develop and ultimately implement new or improved analytical methods and procedures for organic contaminants. Two special projects were undertaken this year:

- Determination of toxaphene in air samples using electron capture negative ion mass spectrometry (ECNI-MS): Efforts have resulted in the successful development of an in-house analytical method for the determination of toxaphene. A feature of this methodology is the calibration approach, based upon technical toxaphene as the available standard, but utilising available information on congener response variability to assign independent response factors to the dominant species seen in ambient air samples. To apply this method to Point Petre air samples, it has been necessary to modify the sampling procedure

to collect larger volumes of air (about 1800 m³). To examine the possibility of vapour-breakthrough on the PUF under these sampling conditions, Supelpak-2 resin absorbent was included in the sampler cartridge downstream of the PUF. Following a brief review of data from mid-summer 1996, a change was made to the sampling procedure in October 1996: results at that time suggested that toxaphene breakthrough on PUF was negligible, even at the maximum summer temperatures, but that significant quantities of PCBs were being found on the resin. While it had been hoped that PUF could serve as a suitable medium for the determination of toxaphene and coplanar PCBs on the same ambient sample, this clearly indicated that this could best be achieved by moving to a sample head utilising only the resin as a vapour trap. These results, however, provided confidence in some earlier toxaphene data for 1992 obtained by sampling at volumes >1000 m³ on PUF. Removal of the PUF also had the advantage of reducing the sample "load" and also reducing wear on the sampler motor by decreasing the flow restriction. Airborne concentrations between 1992 and 1996 appear to have undergone little change, where total concentrations range from a low of 1.8 pg.m⁻³ to a high of 11.9 pg.m⁻³ in 1992 and from 1.1 pg.m⁻³ to 8.1 pg.m⁻³ in 1995/96. In a typical air sample the homolog distribution of the total mass is: octachlorobornane 48%, heptachlorobornane 45% and nonachlorobornane 7%. The chlorobornanes T2 and T12 were found in all air samples but contribute only about 10% of the total toxaphene in air.

- Determination of coplanar PCBs in air samples using multi-dimensional gas chromatography with electron capture detection (MDGC-EC): OAL has developed an analytical technique for coplanar PCBs that could overcome the co-elution and interference issues. In this methodology, two columns of different polarity are connected in series so that the second (main) column can receive selected, well-defined portions of the eluent from the first (monitor) column. This methodology has been previously applied to a set of composited, archived extract fractions derived from selected air samples. The outcome was a very revealing "snapshot" of the seasonal trends and partitioning behaviour. These investigations showed that the target coplanars are present in such background air samples at very low concentrations relative to the "normal" congeners. For the samples initially examined, this difficulty was partially solved by combining the extracts derived from the three replicate samplers operating during this period and reducing the

sample volume down to 200 μl . Even under these conditions, equivalent to ambient air volumes of around 900 m^3 , detectability was found to be marginal for many of the target species, especially for the particle-phase, complicating realistic assessments of gas-phase/particle-phase partitioning. Given current analyzer instrumental detection limits, a doubling of the air volume sampled was deemed a suitable target for improving ambient detectability.

Developed methods for toxaphene and coplanar PCBs provide ambient data at IADN sites

2.2.3 PCBs in Urban Air

- T. Bidleman and T. Harner

Several polychlorinated biphenyl (PCB) congeners elicit toxic responses similar to those of polychlorinated dioxins and furans. A number of these are contaminants of Great Lakes fish and account for the major share of the dioxin-like toxic equivalents (TEQ) in biota and human milk. Some of the polychlorinated naphthalenes (PCNs) also show dioxin-like activity. This report summarizes the results of field measurements and physicochemical properties determinations carried out between 1995 and 1997. The objectives have been to develop analytical methods, to determine the levels in Chicago air, and to compare the particle/gas distributions of PCBs and the sorption of PCBs to other classes of aromatic compounds.

The Junge-Pankow adsorption model is commonly used to predict particle/gas distribution from the specific surface of aerosols and the vapour pressure of the compound. An alternative model was developed which considers absorption of gases into an organic liquid film on the aerosol. The model uses the octanol-air partition coefficient (K_{oa}) of the compound rather than vapour pressure, and the organic carbon fraction of the aerosol rather than its surface area (Finizio et al., 1997). Comparison of the K_{oa} and Junge-Pankow models shows agreement within a factor of two or better for estimating fractions of PCBs and PAHs (polycyclic aromatic hydrocarbons) sorbed to particles (Bidleman and Harner, 1997).

As a descriptor of particle/gas partitioning, K_{oa} is able to resolve the observed differences among the ortho-chlorine classes of PCBs in Chicago that are not explained by vapour pressure. Values of K_{oa} were determined for several PCN congeners as a function of temperature and used to interpret particle/gas partitioning of PCNs in Chicago air. Particulate fractions of PCNs showed a stepwise increase with homolog number, from trichloro- to octachloronaphthalenes. Values of $\log K_p$ were excellently correlated with $\log K_{oa}$.

Two models compare closely for predicting particle/gas distributions in urban air

The sorption of coplanar PCBs to atmospheric particles is enhanced compared to other PCBs of the same homolog group. As a consequence, precipitation scavenging and particle dry deposition are likely to preferentially deliver the most toxic of the PCB congeners to the Great Lakes. A model based on K_{oa} and organic carbon fraction of the aerosol is easier to apply than the conventional Junge-Pankow model based on vapour pressure and aerosol surface area. The two models compare closely for predicting particle/gas distributions in urban air. K_{oa} is able to resolve class differences among the PCBs that are not explained by vapour pressure.

2.2.4 Chlorinated Pesticides and the Great Lakes

- T. Bidleman, T. Harner, L. Jantunen, J. Wideman and K. Wiberg

The project was undertaken with the collaboration of R. Falconer (Youngstown State University), W. Parkhurst (Tennessee Valley Authority), H. Alegria (University of South Carolina), A. Finizio (University of Molise) and S. Szeto (Agriculture and Agri-Food Canada).

Although most organochlorine (OC) pesticides have been banned in Canada and the U.S., they continue to be found in air and precipitation. Sources may include volatilization of soil residues and atmospheric transport of freshly applied material in countries where usage is still permitted, such as Mexico and Central America. This project investigated the present-day sources of OC pesticides to the Great Lakes by measuring concentrations of airborne pesticides, by determining the potential for agricultural soils in the U.S. to contribute OC pesticides to the atmosphere through volatilization, and by investigating the use of marker compounds to distinguish "old" (soil derived) and "new" (freshly applied) sources of OC pesticides.

Toxaphene in ambient air in South Carolina an order of magnitude higher than levels in Great Lakes region

In August 1994, a project was initiated to measure atmospheric concentrations of OCs in the southern states and examine agricultural soils for OCs. It was found that toxaphene in the ambient air of Columbia, South Carolina, averaged 195 $\text{pg}\cdot\text{m}^{-3}$ from August - January (Bidleman et al., 1997), an order of magnitude higher than levels reported in the Great Lakes region. Air sampling was carried out at the

Tennessee Valley Authority (TVA) headquarters in Muscle Shoals, Alabama, from January 1996 to November 1997. Approximately 40 soil samples were obtained from cotton fields and from agricultural research stations in Alabama. Soils showed toxaphene levels ranging from 3-2800 ng.g⁻¹ dry wt, with a geometric mean of 149 ng.g⁻¹. Toxaphene in Alabama air varied seasonally from 134 pg.m⁻³ in winter to 282 pg.m⁻³ in June. Air and water samples were collected from Lake Superior on a cruise of the Limnos in August 1996. Mean concentrations of toxaphene were 28 pg.m⁻³ in air and 800 pg.l⁻¹ in water. Thus the levels of toxaphene in air from Alabama and South Carolina are far higher than over Lake Superior, implying that atmospheric transport is a continuing source of contamination.

Pesticide residues in soils of the U.S. "Cornbelt" are being determined through sampling of approximately 40 agricultural soils in Ohio, Indiana and Illinois during 1995-96. Concentrations of total DDTs were highly variable, ranging from <1-300 ng.g⁻¹ dry wt. Several soils contained dieldrin in the range of 10-110 ng.g⁻¹, and one soil contained over 1500 ng.g⁻¹. Other abundant OCs in the soils were chlordane and heptachlor epoxide, a metabolite of heptachlor. A fugacity-based model is being developed to estimate the volatilization of pesticides from soils. This will be applied to the Alabama and Corn Belt soils, using soil properties which are typical of the regions.

Another thrust of this project was to investigate the use of pesticide enantiomers as tracers of air-surface exchange. Several OC pesticides are chiral ("handed") and are manufactured as mixtures of two enantiomers that occur in a 1:1 proportion (racemic). Selective breakdown of one enantiomer by microbial action in soil and water leads to a residue having an enantiomer ratio (ER) different from 1.0 (Falconer et al., 1997; Ridal et al., 1997). Volatilization of the pesticide can be followed by the appearance of the distinctive enantiomeric signature in air. This was shown by an experiment to measure volatilization at a vegetable farm in the Fraser Valley, BC. Concentrations of α -HCH, chlordanes, o,p'-DDT, heptachlor and heptachlor epoxide in air decreased with height above the soil, and the enantiomeric profiles in the soil were reflected in the overlying air. Non-racemic chlordane, heptachlor epoxide and o,p'-DDT were also found in soils and overlying air at farms in the Corn Belt region.

Volatilization of α -HCH can be traced by appearance of non-racemic α -HCH in boundary-layer air

The α -HCH in Great Lakes waters is non-racemic, with ER values between 0.80-0.85. During spring, fall and winter, the ER of α -HCH in air is racemic, indicating atmospheric transport of the pesticide from continental sources. During

the summer, the water/air fugacity gradients predict volatilization from the lakes by the appearance of non-racemic α -HCH in the air boundary layer. ER values above the lake in summer drop to 0.90-0.95 over Lake Ontario (Ridal et al., 1997) and Lake Superior. Non-racemic signatures are also seen for chlordanes in the air above Lake Superior. It is clear that "recycling" of pesticides from soil and water is contributing to atmospheric levels.

OC pesticides appear to emanate from a variety of sources. The high concentrations of DDTs, aldrin and dieldrin found in Belize suggest that these chemicals are still used in Central America and contribute to the atmospheric background over North America. Toxaphene concentrations in ambient air of the southern U.S. are much higher than Great Lakes values, implying continuous emissions from soils in the region. Volatilization of chlordane, heptachlor epoxide and dieldrin from soils in the Corn Belt may contribute to the regional background. Distinctive enantiomeric signatures have been found for OC pesticides in agricultural soils and Great Lakes waters. These signatures are useful as tracers of volatilization and for distinguishing "recycled" pesticides from fresh sources.

2.2.5 Toxaphene and HCHs in the Arctic

- T. Bidleman, L. Jantunen and T. Harner

This is the final year of a project supported since 1992 by the Northern Contaminants Program. Collaboration occurred with W. Strachan (NWRI), R. Macdonald (IOS) and H. Kylin (Swedish University of Agricultural Sciences, Uppsala, Sweden). The objectives were to measure the concentrations of chlorobornanes (CHBs) and hexachlorocyclohexanes (HCHs) in arctic air and water for comparing the air-sea gas exchange rates of the two pesticides, to determine the physicochemical properties of CHBs that affect their atmospheric transport and deposition, and to characterize the changes in OCs (organochlorines) that accompany transfer from air-water-food chain.

The research comprised of four field studies: Resolute Bay (1992); Bering-Chukchi seas (1993); Arctic Ocean Sections cruise (1994); and Barents Sea and eastern Arctic (1996). Two subchapters were written for the Canadian Arctic Contaminants Assessment Report (CACAR): one on the mass balance of HCHs in the Arctic Ocean and the other on the atmospheric budget for CHBs.

A surprising discovery is that the OC distribution in arctic surface water is not uniform. The sum of HCHs is 4-5 ng.l⁻¹ in the central Canada Basin, the Beaufort Sea and the Archipelago, 2.5 ng.l⁻¹ in the Bering and Chukchi Seas and across the northern Canada Basin, and less than 1 ng.l⁻¹ in the Greenland Sea, the Barents Sea and the eastern Arctic Ocean. CHBs and heptachlor epoxide are also higher in the Canada Basin than in the Bering-Chukchi seas.

Reversal of the net gas exchange direction from deposition in 80s to volatilization in 90s.

Due to declining atmospheric concentrations, the gas exchange flux of α -HCH in the Canada Basin and the Bering-Chukchi seas has reversed from deposition in the 1980s to volatilization in the 1990s. The greatest potential for outgassing is in the Canada Basin, but the actual loss of α -HCH is limited by ice cover. Water concentrations of α -HCH are much lower in the eastern Arctic and gas fluxes in this region are still depositional.

Enantiomers selectively degraded in arctic waters

The two enantiomers of α -HCH are broken down at different rates in arctic waters. Selective degradation of (+) α -HCH occurs in the Canada Basin and (-) α -HCH in the Bering-Chukchi seas. The opposite selectivity may result from different microbial populations in these waters. The air sampled within 40 m of the ocean surface shows the same profile of α -HCH enantiomers as the surface water. This demonstrates that gas exchange is indeed a "two-way street" and that revolatilization from the ocean surface contributes HCH to boundary-layer air (Jantunen and Bidleman, 1996).

A mass budget for HCHs in the upper 200 m of the Arctic Ocean was constructed from ocean current advection, riverine input and atmospheric processes. Ocean currents provide 76% of the HCH input, with atmospheric deposition and rivers contributing 22% and 2%, respectively. Outflow of HCHs takes place mainly through the Canadian Archipelago (46%) and the East Greenland Current (22%). Volatilization and ice export account for 16% and 2% of the losses. The only chemical loss quantified was hydrolysis, which was estimated to account for 14% of HCH removal. However, microbial breakdown is evident from the enantiomeric patterns of α -HCH in the water column and this process may be a more important sink.

A scenario for CHBs indicates that the gas exchange flux is depositional on an annual basis and that most of the exchange takes place during the summer months when atmospheric concentrations are highest and the area of open water is maximal. Precipitation and dry deposition of CHBs contribute more substantially than for HCHs because of the greater fraction of CHBs associated with haze aerosols.

Concentrations of CHBs and HCHs are higher in the Canada Basin than in other arctic waters. This pool of enriched pesticides may be a remnant of higher atmospheric deposition in the past, coupled with more limited removal by

revolatilization and sedimentation of particles under the ice pack. The outflow of polar water containing elevated concentrations of OCs may impact biota in the Canadian Archipelago for years to come. This is the first report of microbial breakdown of an OC pesticide in arctic water, and has significance for loss processes in the polar environment.

2.2.6 Smog Chamber Studies

- D.A. Lane

The study of the reaction of the OH radical with the polycyclic aromatic hydrocarbon, naphthalene, has continued. The research now has both an experimental and a theoretical component to it. The experimental work is being done in collaboration with Dr. N. Bunce (U. Guelph), while the theoretical work is being done by Dr. S. Townsend (Genexus Inc.) and Dr. S. Fielder (Central University of Queensland, Rockhampton, Australia).

During the past year, the two mass spectra characterized by the $m/z=131$ mass peaks have definitively been identified. They were the (E) and (Z) forms of 2-formylcinnamaldehyde. Reaction mechanisms were worked out to explain why the 1- and 2-naphthols and the 1- and 2-nitronaphthalenes are not on the major decomposition pathway. The well-known mechanism of Atkinson suitably accounts for these minor pathway products but does not account for the vast number of products isolated in our experiments both by GC/MS and by HPLC. The mechanism involves attack by oxygen after the initial attack by the OH radical to form a peroxy radical rather than rearomatization. From the peroxy radical, the products seen by the GC/MS can now be accounted for.

Suitable reaction mechanisms have been elucidated for the following products: 1- and 2-naphthol, 1- and 2-nitronaphthalene, 2-nitro-1-naphthol, 4-nitro-1-naphthol, 1-nitro-2-naphthol, naphthoquinone, (Z)- and (E)-2-formylcinnamic acid, 2-formylcinnamic acid and the 2,3-epoxynaphthoquinone. Products indicated by HPLC include oxalic acid and benzoic acid.

A contract with Genexus Inc. was established to use the capabilities of the Frontier Molecular Orbital (FMO) Mapping program (a generalized form of the Graphic Electronic States Molecular Modeling (GESMM) program developed by Dr. S. Fielder) to model the reaction of naphthalene with the OH radical. The ultimate objective of the work is to use the GESMM in a predictive manner to indicate what products ought to be found when an organic compound (PAH) reacts with the OH radical (or, in principle, any oxidant). A report submitted by Genexus Inc. is currently under review.

2.2.7 Denuder Development

- D.A. Lane

For the past three years, through collaboration with Dr. L. Gundel of the Lawrence Berkeley National Laboratory (LBNL), the engineering design of the GAP sampler (developed by ORTECH and AES) was merged with the surface coating procedure developed by Dr. Gundel for the Integrated Organic Vapor and Particle Sampler (IOVPS). The new sampler, appropriately called the Integrated Organic Gas and Particle Sampler (IOGAPS), is now commercially available from University Research Glassware in Chapel Hill, NC. The first commercial unit has been modified to withstand Arctic temperatures.

Jumbo IOGAPS that samples air at a rate of 100 L/min through each of two denuders built at LBNL

In July 1996, a workshop held by the US EPA discussed the measurement of dioxins in the environment. The consensus of the attendees (representatives from the USA, Canada, the United Kingdom, Sweden and Germany) was that it was mandatory that gas/particle partition measurements of the dioxins be made. It was also recognized that the technology to accomplish this task was the IOGAPS. It was requested that the IOGAPS be modified to be able to sample 1000 m³ of air in as short a time as possible.

Over a period of 6 weeks, the Jumbo IOGAPS, an instrument that samples air at a rate of 100 L.min⁻¹ through each of two denuders, was engineered and built at LBNL. A conventional PS1 sampler also operates in parallel to the two denuders. In two 24-hour periods, sufficient air should be collected to make a quantitative measurement of the 2,3,7,8-tetrachloro-p-dioxin.

In February 1997, the new IOGAPS and 4 denuders were shipped to LBNL for more collaborative work. The annular diffusion denuders built by URG have a much more uniform gap width than those of the original GAP sampler and the gaps are narrower (most being 1.0 mm wide; the largest being 1.4 mm). Using the particle diffusion equations of Yeh et al. (1991), it was determined that the denuder should have a loss of 2.7% or less for particles greater than 0.1 µm. A laser optical particle counter was used to determine the loss of particles through the denuder. Although there was no detectable loss through the denuder, it should be noted that there was a 3% noise on the signal for particles of 0.09-1.2 µm. The conclusion was that the denuder was functioning just as the theory predicted.

2.2.8 Atmospheric Particulate-Phase Mercury

- J. Lu, W.H. Schroeder and A. Steffen

The project objective was to develop a viable procedure for sampling and analysis of total particulate-phase mercury (TPM) in ambient air in collaboration with D. Schneeberger and F. Schaedlich of TEKRAM Inc., Toronto.

Although it accounts for only a few percent by mass of the total mercury (Hg) occurring in ambient air, mercury in the particulate phase is critical in understanding the cycling of this metal in the environment because, relative to elemental mercury vapor, mercury-containing particles in air are readily deposited to the earth's surface, through rain- and wash-out processes. While methods for the determination of total gaseous-phase mercury (TGM) in the atmosphere have been standardized in recent years, techniques and methods for effectively collecting and accurately analyzing TPM without artifacts are still under development. Traditionally, TPM is determined by collecting the airborne particles using a glass/quartz fiber filter followed by wet digestion of the particles along with the filter. Because of the possible interactions among particles retained, the filter materials and vapor-phase Hg species during sample collection, condensation of vapor-phase Hg onto the particles retained by the filter and/or release of weakly-bound volatile Hg from the particles into the vapor phase may occur. Hence, the results obtained by that technique may be unreliable.

Significant break-through in measurement of atmospheric particulate-phase mercury

A system combining an gold-coated denuder with a particulate collector has been constructed with the denuder performance optimized in terms of length versus flow rate. The collection efficiency of a quartz fiber filter disc, granular magnesium oxide (MgO), quartz wool and vycor beads were tested as packing materials in the particulate collector. The optimized denuder-based system, which will collect TGM first, and a TEKRAM Mercury Vapor Analyzer attached to a particulate collector, which will remove TPM first, will be tested side-by-side for a comparison of the two sampling techniques.

An analytical apparatus for the trace determination of TPM was assembled. Traps consisting of a quartz tube containing either quartz wool, a quartz fiber disc or granular MgO as packing material were made in the laboratory and have been tested for ambient air sampling and the determination of TPM. Equivalent results (relative error of 2%) were obtained using a quartz wool trap and a MgO trap for the determination of mercury concentration in a standard reference material (SRM 1648, NIST). The standard deviation for measurement of

TPM in ambient air, including sampling and analysis, is $<\pm 2 \text{ pg.m}^{-3}$ ($n = 4$).

A pyrolysis technique for the determination of TPM has been validated using an alternate standard reference material (SRM 1633b, NIST), for which a certified value for mercury exists. The value determined in our laboratory and the certified value agree well with a relative error $<5\%$. Pyrolysis analysis has also been tested for chemical speciation of TPM. Results obtained so far show that over 80% of mercury in urban particulate matter is thermally labile at 300°C .

Denuders (60 cm long with a 0.4 cm inner diameter) can remove 98% of the vapor-phase Hg^0 in a spiked air stream ($[\text{TGM}] = 5.0 \text{ ng.m}^{-3}$) in the laboratory at a flow rate of 1.5 l.min^{-1} . Collection efficiency drops to 90% if ambient air is used. In tandem, two such denuders can remove 99.8% of TGM in ambient air at a flow rate of 1.0 l.min^{-1} , even after 52 hours of sampling. TGM determined at the Downsview Field Test Site between May and June 1996 ranged from $3.0\text{-}7.5 \text{ ng.m}^{-3}$ (5 minute sampling time). TPM, collected at the same site and at the same period of time, using the traps described above and analyzed using the pyrolysis technique, ranged from $0.028\text{-}0.090 \text{ ng.m}^{-3}$. These values for TPM fall within the range ($0.001\text{-}0.100 \text{ ng.m}^{-3}$) of those determined elsewhere in the Great Lakes region.

2.2.9 Atmospheric Mercury: Alert

- W. Schroeder, A. Steffen, V. Hudec and L. Barrie

This project obtained data on background ambient air concentrations for total gaseous mercury (TGM) at Alert. These data will be used to assess the current baseline concentrations of TGM in the Canadian High Arctic, to determine the temporal variability of TGM concentrations, to identify regions of major emission sources of mercury, and to determine the significance of long-range atmospheric transport and deposition of this priority heavy metal to Arctic ecosystems.

In 1996, following polar sunrise, it was observed that there was a very interesting temporal pattern of TGM concentrations: the variability of the observed data is much greater, and relatively low ($<1 \text{ ng.m}^{-3}$) ambient air concentrations of TGM occur much more frequently, at Alert during the spring (late March-April-May) than for other times of the year. It is quite remarkable (and totally unexpected) that the TGM pattern strongly resembles that of surface-level ozone which exhibits dramatic depletion events following polar sunrise. In the case of ozone, this phenomenon has been linked to photochemical production of very reactive chemicals which destroy ozone. The reason for the "depletion" of

mercury vapour in the boundary layer of Arctic air during the spring months is not yet known.

Mercury vapour in Arctic air observed to mimic surface-level ozone depletion after polar sunrise

Finally, the earlier atmospheric mercury measurement results from Alert have recently been used to identify possible source regions of atmospheric mercury emissions likely to contribute to the concentrations of this heavy metal observed at Alert. This collaborative work with Oak Ridge National Laboratory (Tennessee, U.S.A.) involved the application of a numerical modelling technique called the "Potential Source Contribution Function (PSCF) source-receptor model (Cheng and Schroeder, 1996).

Diagnostic testing performed on the Tekran analyzer operating at Alert, during late March and early April of 1997, has shown conclusively that the low TGM levels are real and that the possibility of a measurement artifact can be ruled out (viz., passivation of the gold traps which pre-concentrate the mercury in the ambient air sample prior to its analytical determination, or quenching of the fluorescence detector signal by an unknown contaminant).

This work was undertaken in collaboration with D. Schneeberger (Tekran Inc.), G. Vandal, C. Lamborg and W.F. Fitzgerald (U. Connecticut), T. Berg (Norwegian Institute for Air Research, NILU, Oslo, Norway) and S. Junnto (Finnish Meteorological Institute, Helsinki, Finland).

2.2.10 Atmospheric Mercury: Lake Superior

- W.H. Schroeder and A. Steffen

The expedition, August 3-16, 1996, on the ship, CSS Limnos, obtained summer-time air concentration data of total gaseous mercury (TGM) over Lake Superior. This work was done in collaboration with scientists from AES and Canada Centre for Inland Waters (CCIW) who were measuring persistent organic pollutants, and selected metals in both water and air media. The atmospheric mercury concentrations were obtained using a Tekran mercury vapour analyzer. Collaborators on this project were W. Strachan and G. Lawson (CCIW), and G. Eiras (TEKRAN Inc.). Testing was also undertaken on this cruise, jointly with G. Lawson of CCIW, to analyze dissolved gaseous mercury in various water samples.

The mercury analyzer was operated at five minute integrated sampling intervals for 24 hours a day. It was located in the chart room adjacent to the bridge on the

upper deck of the ship. A 15.25 m heated Teflon sample line, with a 47 mm diameter Teflon (0.2 mm) particulate filter at the inlet to trap any particulate matter, conveyed ambient air from the top of the mast to the analyzer.

The TGM concentrations for the first 4 days were quite constant and consistent with expected values, ranging from approximately 1.9-2.5 ng.m⁻³. These data cover the southeastern and northeastern parts of Lake Superior. The morning of August 8, a pressure gauge (mercury manometer) broke in the engine room and resulted in a liquid mercury spill. Following the spill, the mercury levels detected by the analyzer rose from approximately 3 ng.m⁻³ to 25 ng.m⁻³ and remained high for the remainder of the expedition.

The first three days of the voyage yielded good results for TGM concentrations over Lake Superior. Because of the mercury spill on board the ship and an instrument leak, data obtained for the remainder of the expedition are not representative of Lake Superior air masses. Table 1 summarizes the daily averages, the median and the ranges of mercury concentrations during the first three days of the Lake Superior segment of the expedition.

Table 1: Total Gaseous Mercury Concentrations (ng.m⁻³)

	Daily Average	Median	Range
5-Aug	2.19	2.17	2.03-2.40
6-Aug	2.24	2.23	2.09-2.29
7-Aug	2.26	2.19	2.05-3.30

As well on this expedition, a preliminary study into determining the concentration of dissolved gaseous mercury (DGM) in various water samples was investigated. Samples were collected in a 4l Go-Flo bottle and transferred to a 1l Teflon bottle. The samples were sparged with argon and air (at different times). The purged gas stream was fed through an in-line Nafion drying tube and then to the Tekran mercury vapour analyzer for detection. These preliminary studies showed that this technique could be a viable means of determining the DGM available for air-water exchange processes.

2.2.11 Air-Soil Exchange of Mercury

- W. Schroeder and A. Steffen

A 2-week pilot field study during the summer of 1996 tested prototype equipment over mercury-enriched soils at the Clyde Forks mineral deposit in southeastern Ontario (about 100 km southwest of Ottawa). This study also looked at the possibility of observing the temporal and spatial variability of volatile gaseous mercury fluxes at that site by application of the 2 methods available for

making flux measurements of gaseous/volatile mercury emissions from soil (and water) surfaces.

At present, two distinct methods for determining fluxes of trace atmospheric gases, including volatile mercury (Hg) species, over soil (or water) surfaces are theoretically practicable: enclosure-based methods and micro-meteorological methods. From July 8 to July 23, 1996, two dynamic flux chambers were operated in the field at Clyde Forks, Ontario, at sites for which the Geological Survey of Canada (P. Rasmussen) had determined the total Hg content of the surficial soil horizon. The first chamber, constructed from a transparent plastic (Plexiglass), was designed by the Atmospheric Environment Service, while the second (smaller) chamber, fashioned from Teflon film supported by an external frame, was similar to that currently being utilized by the Oak Ridge National Laboratory (ORNL, Tennessee, U.S.A.) for their flux measurements over soil and water surfaces (S.E. Lindberg). Additionally, a micro-meteorological (eddy correlation) flux measurement method was evaluated concurrently at the same field study site by G. Edwards (U. Guelph). The common element of the 2 different (but complementary) methods was their reliance on Tekran mercury vapour analyzers for determining total gaseous mercury (TGM) concentrations in the ambient air at 2 heights above the surface, and in the air streams entering and leaving the 2 flux chambers.

AES prototype flux chamber evaluated for measurements of air-surface exchange fluxes

Each flux chamber was operated primarily in the dynamic mode (usually with auxiliary air flow) during this study; however, a number of static runs were also completed. The Clyde Forks setting allowed for testing and evaluating the equipment under both high and low air-soil Hg flux conditions, thereby facilitating the evaluation of the dynamic range capabilities of the chambers deployed. Furthermore, experiments were completed to investigate the effect of varying the total flow-rate of ambient air through the flux chambers.

Soil emission fluxes of Hg tended to reach the daily maximum values around mid-day. The AES flux chamber yielded mean hourly Hg flux values up to approximately 1500 ng.m⁻².h⁻¹ on sunny days at the high Hg site (# 2) located near the geological fault line. The site (#1), situated directly on the geological fault line, exhibited mean hourly Hg emission fluxes up to about 6500 ng.m⁻².h⁻¹ between 1300h and 1400h on July 12. The agreement between the 2 flux chambers ranged from excellent to poor over the duration of the field study.

Possible reasons for disagreement include high small-scale spatial heterogeneity of Hg-in-soil concentrations, difficulties with obtaining good seals around the flux chambers and operation under non-optimal conditions.

Experiments to determine the potential effect of auxiliary ambient air flow rate on the fluxes measured with the 2 chambers was inconclusive: if there is an effect, it is probably not significant in most flux chamber measurement situations. Soil emission fluxes of Hg consistently followed a diurnal trend and appeared to be directly correlated with the magnitude of net solar radiation ($W.m^{-2}$) measured at the study site. Furthermore, data suggested that emission fluxes are probably influenced by ambient air temperature, and perhaps also by top soil temperature as well as humidity.

Soil emission Hg fluxes consistently followed a diurnal trend and correlated to net solar radiation

The highest emission fluxes were measured at sites #1, #2 and #3, which also corresponded to the locations with the highest total Hg concentrations in the soil. At these 3 sites, emission fluxes overnight were low, but always corresponded to emission (positive) fluxes. Negative fluxes (corresponding to deposition of gaseous Hg) occurred only during the night-time at site #4 which is further away from the geological fault line (the "natural Hg hot spot") than the 3 other sites.

Field testing and in-field operation/evaluation of two prototype flux chambers were successfully completed during July 1996. This work sets the stage for future collaborative measurements of mercury emissions from various types of soils, or different geological "provinces" or features represented within Canadian ecosystems/landscapes, with a view towards a better definition of the magnitude and importance of natural emission sources of mercury in the Canadian (and possibly North American) context.

This study included collaboration with G. Edwards (U. Guelph), P. Rasmussen (Geological Survey of Canada), and G. Eiras and D. Schneeberger (Tekran Inc.).

2.2.12 Harmonization of Buffers in North America - R.E. Mickle

Within the NAFTA (North America Trade Agreement) Technical Working Group on Pesticides, Canada and the USA agreed to collaborate on developing a unified approach to minimize environmental insult from pesticide use. As part of the collaborative effort, Canada agreed to participate in a review of the US Spray Drift

Task Force (SDTF) database on off-target deposit from application strategies involving ground, orchard air-blast and aerial treatments. The review has been forwarded to Pest Management Regulatory Agency (PMRA), Canada's representative on the NAFTA Working Group. Coincidental with this review, AES has actively participated in the SDTF sub-committee that is developing a model to predict off-target deposit for different application strategies. This model extends the approach used in Canada for buffer zone assessment of aerial sprays to include ground and air-blast spraying through the use of representative curves of the SDTF database. In March, a training session was held at PMRA to introduce regulatory staff to the SDTF model. The generic approach brings a strong scientific basis to the registration process for the development of protective strategies for minimizing environmental impacts from pesticide use. With the transfer of the approach to PMRA, this component of pesticide research has been completed within AES.

Approach brings scientific basis to registration process for development of protective strategies

2.2.13 Air-Water Interface Studies - A. Lo

A method for evaluating dry deposition velocities of particulate aerosols has been developed based on the direct method of Lo (1993) which requires only a single level data for determining boundary layer flux parameters. This study considers the particle growth due to humidity effect in a thin deposition layer at the air-water interface, the effects of broken water surface due to white caps, spray and bubbles and effects of meteorological parameters in the turbulence transport layer within the constant flux layer regime. Results of the present study are compared against previous studies that ignored some of the meteorological factors that have now been taken into account in this study. The present study shows that atmospheric humidity, which plays an important role on the atmospheric stratification (Lo, 1996), has a significant effect on deposition velocity of aerosol particles with dry diameters between 0.1-1 μm .

Atmospheric humidity has a significant effect on deposition velocity of aerosol particles

An explicit method for determining drag and heat transfer coefficients at an air-water interface has been developed. Calculations were carried out based on a single level of wind and temperature in addition to the

water surface temperature. Roughness length for momentum was based on a formula that composed of both the Charnoch formula and a term representing an aerodynamically smooth surface. The roughness length for heat is calculated as a function of roughness Reynolds number. Transfer coefficients as functions of wind speeds and air water temperature differences calculated using the present method compared favourably against four other commonly known methods by Kondo (1975), Large and Pond (1982), Liu and Schwab (1987), and Smith (1988).

2.2.14 Air Pollution Prediction System

- S.M. Daggupaty and P.C. Cheung

Development has been completed on combining BLFMESO (Boundary Layer Forecast Mesoscale Model) with an air pollution prediction system. The Boundary Layer Forecast Mesoscale Air Pollution Prediction Model (BLFMAPS) consists of three numerical models: BLFMESO, a trajectory model, and a dispersion/deposition model. The system was designed for Pickering, Darlington and Bruce nuclear power facilities. The trajectory model uses the variable acceleration algorithm (Daggupaty, 1980) which is an essential technique for studying transport of pollutants in the boundary layer. The dispersion model is a multiple puff hybrid model. The 3D trajectories of puffs and peak concentration in the air along each trajectory are used to calculate concentration as a function of time at nearby grid points in the Eulerian frame. The trajectory and dispersion algorithms use BLFMESO predicted parameters and modified Hefter's mesoscale horizontal diffusion coefficient as a function of time. Dry deposition is parameterised using the land-use category of the domain and pollutant type. Wet deposition is accounted for by using the CMC predicted precipitation rate at each grid point. The system accounts for radionuclides of importance from CANDU reactors. With our modelling system it has been shown that the pollutant transport and the concentration variation diurnally and day to day are significantly influenced by the mesoscale processes. The BLFMAPS has been implemented in AECSB (Atomic Energy Control Board) Ottawa office.

2.2.15 Global Emissions Inventories

- Y-F Li

The relationship between the global technical HCH use trends and their impact on the arctic and sub-arctic atmospheric environment has been studied. Two significant drops of global α -HCH usage were identified. One was when China banned the use of technical HCH in 1983, and another occurred around 1990 when both India and the former USSR also banned the usage of technical HCH. From measurements of air concentrations of α -

HCH in arctic and subarctic regions from 1979 to 1994 (Bidleman et al. 1995, Jantunen and Bidleman, 1995, 1996), the long-term trends also showed two significant decreases, one happened between 1982 and 1983, and another between 1990 and 1992.

The consistency between these data shows direct connections between global usage of α -HCH and the air concentration of α -HCH in the arctic, as evidenced by the reductions of air concentrations of α -HCH in arctic and subarctic regions corresponding to reduction of global α -HCH usage.

Direct connection observed between global usage of α -HCH and air concentration of α -HCH in Arctic

2.2.16 Air Pollution Initiatives

- S.M. Daggupaty

S.M. Daggupaty participated as a working group member of the Hamilton-Wentworth Air Quality Initiative in connection with the air quality of Hamilton and its impact on the environment. A comprehensive report, dealing with local emission of hazardous pollutants, ambient concentrations, trends and impacts, will be published soon. The program is led by Ontario Ministry of Environment and Energy.

Licensing agreement with Environmental Software Consultants, with a royalty for the Government of Canada, was completed for the advanced MS Windows 95 version of PC-AQPAC (Air Quality Package for environmental emergencies) now known as ASAP (Air Safety Applications Package for emergency response). Training was given to Thunder Bay regional weather office meteorologists on the use of PC-AQPAC model particularly with regards to theory, special applications and validation results.

2.3 TROPOSPHERIC OZONE

2.3.1 CAPMoN Ozone and NO_x Measurements

- D. MacTavish, N. Lance, R. Braga, B. Kessler, B. Kiely, W. Kobelka and D. Ord

CAPMoN (Canadian Air and Precipitation Monitoring Network) continues to support the NO_x/VOC (Nitrogen Oxides/Volatile Organic Compounds) program through its network air filter and precipitation measurement programs by determining concentrations of nitrogen compounds in air and precipitation samples.

Continuous ozone measurements were maintained at 8 of CAPMoN's monitoring sites and NO_y measurements at 1

site. Operations were maintained with the help of the DOE regional staff. Quarterly inspections/calibrations were performed at all 8 ozone measurement sites. The year 1996 marked the third year of a intensive ozone audit program designed to ensure the accuracy and quality of CAPMoN's ozone data.

Development work continues on the TECO model 42S NO_x analyser. This instrument operated in an NO_y mode at Egbert during 1996. Operational protocols are still under development.

A significant portion of CAPMoN resources were committed in support of the 1996 NARSTO-CE (North American Research Strategy for Tropospheric Ozone-Canada East) field study. CAPMoN's technical staff designed and implemented 75% of the air filter measurements equipment required while the laboratory analysed over 8000 filter samples and reported in excess of 24,000 results.

2.3.2 Differential Absorption Lidar Project

- R.M. Hoff and A. Sheppard

Over the past few years, Differential Absorption Lidar (DIAL) remote-sensing techniques have been developed to profile ozone up to the tropopause. Unlike stratospheric ozone, which can (and must) be probed using wavelengths above 308 nm, tropospheric ozone is found at much smaller concentrations in the atmosphere and requires use of wavelengths further into the ultraviolet. In addition, tropospheric aerosols are a major confounder in the retrieval of ozone profiles from DIAL. A recently-constructed tropospheric ozone DIAL, based on two Nd:Yag lasers which pump visible dye lasers, is now being tested. The output of each laser being doubled to wavelengths near 290 and 295 nm. From the differences on these two wavelengths, ozone concentration in 150 m vertical range cells can be determined.

DIAL remotely senses vertical profiles of tropospheric ozone for the first time in Canada

In March 1997, profiles to 5 km altitude were retrieved on the smaller of two telescopes dedicated to the system. When the larger 40 cm telescope is operational, early in the summer of 1997, ozone profiles into the lower stratosphere will be able to be retrieved. The intent of the project is to study tropospheric/stratospheric exchange processes. This project complements studies of stratospheric ozone made by DIAL at ISTS/York University (Dr. A. Carswell) and profiles of water vapour

being made at University of Western Ontario (Dr. R. Sica).

2.3.3 QUEBEC'96 - Mt. Sutton

- K. Anlauf, A.M. Macdonald, M. Watt, K. Hayden, D. MacTavish and P. Makar

Results from this study will aid Eulerian model evaluation, NARSTO (North American Research Strategy for Tropospheric Ozone) participation for large-scale geographical database, and photochemistry investigations at high elevation (day versus night). Collaboration was obtained from D. Wang and T. Dann, EPS.

NARSTO is a cooperative effort involving national and regional governments, industry and academia in the United States, Canada and Mexico to investigate the tropospheric ozone problem. The Atmospheric Environment Service (AES) participated as part of NARSTO-CE (NARSTO-Canada East) in an intensive field campaign that was carried out in the summer of 1996. Measurements taken at two sites in Quebec (Mt. Sutton and St. Anicet) and one site in Nova Scotia (Kejimikujik) consisted of two phases: a 'low intensive' sampling period from mid-May to mid-September where basic chemistry/meteorological measurements were made; and a 'high intensive' sampling period from mid-June to mid-July where more detailed chemistry/meteorological measurements were included.

The Mt. Sutton, Quebec, site (845 msl, 90 km southeast of Montreal) is located at high enough elevation so as to be in the planetary boundary layer, thus this station intercepts long-range transport of pollution throughout the day. It provides a unique opportunity to study chemical interactions without the complication of a nocturnal boundary layer. Extensive measurements of ozone (O₃), nitrogen oxides (NO_y, NO₂, NO, PAN, HNO₃) and volatile organic compounds (VOCs) were made in order to characterize and understand the O₃-NO_x-VOC relationships in this region. Other measured compounds important in oxidant transport and chemistry included HCHO, H₂O₂, particles, CO, NH₃, organic acids and a suite of meteorological parameters.

Preliminary results from Mt. Sutton presented at a workshop in December 1996

Detailed chemistry and meteorological measurements were made at Mt. Sutton from mid-June through mid-July 1996 with preliminary results being presented at a workshop in December 1996. The results indicated that:

- the site was frequently impacted by cloud and precipitation leading to reduced levels of HCHO and H₂O₂;
- solar radiation was the lowest in the past 10 years for June and July resulting in minimal local photochemical activity;
- the mean O₃ was 37 ppbv and was typical of levels observed at this time of year as ascertained from previous 10 years of measurements;
- a formaldehyde maximum correlated with a maxima in temperature, solar radiation, isoprene and hydrogen peroxide;
- NO_y was always <4 ppbv, most of the time <2 ppbv and consisted mostly of PAN and HNO₃ in similar proportions; NO was always <1.5 ppbv;
- HNO₃ during periods of pollution transport was 0.5-1 ppbv with several peaks up to 2 ppbv;
- PAN concentrations on average were 0.2-0.8 ppbv; a maximum was reached of ~1-1.5 ppbv and a minimum of <0.1 ppbv;
- p-NO₃⁻ was a smaller NO_y constituent and correlated with elevated pollution concentrations; and
- particles measured simultaneously at Mt. Sutton and St. Anicet during the arrival of a moderately polluted (O₃ ~50 ppbv) air mass showed similar characteristics to particles observed during a pollution event at Egbert, Ontario, in 1992.

2.3.4 Sensitivity of Heterogeneous Processes

- H.A. Wiebe, R. Staebler and S.M. Li

Particles in polluted airmass at night control nitric acid chemistry, and impact on acid deposition

To determine the sensitivity of gas-phase chemistry to heterogeneous interactions with particulate ammonium nitrate, a particle nitrate model was constructed and coupled to an existing gas-phase chemistry model. A large number of simulations with the new model were used to estimate the effects of particle nitrate formation on gas-phase chemistry. The model was then used to simulate observations of acidic particle formation made at the Egbert (CARE) site in August of 1992. Papers have been written that describe the sensitivity study and the comparisons to measurements.

2.3.5 Gas-Phase Chemistry

- P. Makar, S-M. Li and S. Polavarapu

The reaction mechanism currently in use in the ADOM-II, MC2-CTM and MC2/ADOM models was last updated in 1988. Laboratory experiments carried out subsequent to that time have greatly increased our knowledge of

atmospheric chemical processes. This project has the aim of incorporating this new information into a revised chemical mechanism for regional modelling.

Hydrocarbons combined in one variable to reduce pollution prediction computation time

A paper has been published that examines the mathematical methods used for combining hydrocarbons in gas-phase chemical mechanisms, and describes a new method by which many hydrocarbons can be combined as a single variable with no loss in accuracy to the rest of the chemical system. In another paper, the current ADOM-II mechanism was compared to 11 other reaction mechanisms for several test cases.

As part of the EEC Eurotrac project, the following collaborated: M. Kuhn, P.J.H. Builtjes, D. Poppe, D. Simpson, W.R. Stockwell, Y. Andersson-Skold, A. Baart, M. Das, F. Fiedler, O. Hov, F. Kirchner, P.A. Makar, J.B. Milford, M.G.M. Roemer, R. Ruhnke, A. Strand, B. Vogel and H. Vogel.

2.3.6 Gas-Phase Chemistry Box Model

- S.M. Li and P. Makar

An advanced Windows 95 front-end code has been constructed and coupled to a personal computer version of the model used by P. Makar for chemistry simulations. The new program will allow scientists not familiar with modelling to make use of an advanced simulation tool in conjunction with their measurement/monitoring research. The Windows 95 front end was created by J. Guilarte. The "Alpha" version of the code is currently being tested/debugged. The "Beta" version should be ready for internal AES users to test out in the early summer of 1997.

Windows-based chemistry model makes complex gas-phase chemistry simulation easily accessible

2.3.7 Mesoscale Boundary Layer Forecast Model

- S.M. Daggupaty

Collaborative work with NYSDEC (New York State Department of Environmental Conservation) has resulted in a study with the analysis and interpretation of monitored ozone with the simulated meteorology from the Mesoscale Boundary Layer Forecast Model (BLFMESO). The study used NARSTO-NE (North American Research Strategy for Tropospheric Ozone-Northeast) field study data. A paper was presented in the

NARSTO-NE workshop in Norfolk, VA. The ozone episode during July 1995 over the northeastern USA was explained with the use of simulated boundary layer flow, mixed layer height and vertical velocity. It concluded that detailed boundary layer processes including sea breeze over this complex terrain are essential for predicting ozone episode.

2.3.8 ADOM/MC2 Modelling of Regional Oxidants

- W. Gong

To enhance the understanding and modelling capabilities of regional oxidants, modelling work continued on the ADOM/MC2 model. This year the focus was on further model evaluation and a modelling-based O₃-NO_x-VOC sensitivity study. In addition to the existing simulation of the EMEFS-1 (Eulerian Model Evaluation Field Study) intensive period, extended model runs were carried out for further model evaluation, as well as to serve as a basis for the O₃-NO_x-VOC sensitivity analysis (through the analysis of indicator species).

A three-month simulation, from June 1 to August 31, 1996, which corresponded with the NARSTO-CE (North American Research Strategy for Tropospheric Ozone-Canada East) study period, was conducted making use of the forecasted meteorological fields from the real-time MC2 run during the same period. Some preliminary comparisons with the observations, for example, the ozonesonde at Kejimikujik, were carried out. A full evaluation will be pursued when the complete data set becomes available.

Trajectory analysis based on the existing model simulation of the EMEFS-1 intensive period was performed to elucidate the role and mechanisms of transport and transformation affecting the South Atlantic Region (SAR) ozone. The study suggests that the high ozone experienced in SAR during the modelled period is related to transport at very low levels and that transport time, as well as timing, are critical for the SAR ozone event.

2.3.9 ARCTOC 1996

- R.M. Staebler, D. Toom, L.A. Barrie, S.-M. Li and J. Bottenheim

Tropospheric ozone depletion processes examined in the European Arctic

The ARCTic Tropospheric Ozone Chemistry (ARCTOC) project is the European equivalent of our Polar Sunrise Experiments, designed to shed more light on the processes involved in the episodic depletion of

tropospheric ozone in the arctic spring. The Branch participated in the second field component of the 2-year study, from 23 March to 14 May 1996, in Ny Ålesund (79° N, 12° E) on the island of Spitsbergen, Norway, in collaboration with the University of Heidelberg and other institutes from Germany, Norway, Denmark, France and the UK. The three main objectives were: a) to determine the physical size characteristics of the aerosols in arctic haze at Spitsbergen; b) to speciate inorganic gaseous and particulate bromine and nitrogen compounds to diagnose mechanisms for ozone destruction; and c) to compare these aspects of arctic tropospheric chemistry at Spitsbergen with those at Alert.

First results show aerosol concentrations and size distributions quite similar to those found at Alert

First results show aerosol concentrations and size distributions quite similar to those found at Alert. Chemically, the aerosol was dominated by sulphate and sea salt components. Increases of both particulate and gaseous bromide during low ozone episodes were confirmed, although concentration levels were significantly below those observed at Alert.

2.3.10 RDMQ System

- W. Sukloff and R.J. Vet

This Research Data Management and Quality Control (RDMQ) system is used in all the program areas of the Branch but this year concentrated on the NO_x/VOC area. The NARSTO-CE (North American Research Strategy for Tropospheric Ozone-Canada East) summer field study presented an opportunity to use RDMQ to quality control data other than the usual long term monitoring data. In NARSTO-CE, a "level 0" quality control module was created to present immediate feedback to scientists in the field in the form of time series plots. The data were read into RDMQ, where a number of scientists took advantage of the capability of quality controlling and applying calibrations.

RDMQ was implemented for the mercury monitoring program in the DOE Atlantic Region. Other AEP groups monitoring mercury are considering using RDMQ, based on the experience in the Atlantic Region.

United States EPA expand use of RDMQ to include the Lake Michigan Mass Balance project

The United States Environmental Protection Agency expanded the use of RDMQ to include the Lake

Michigan Mass Balance project. Development for this under the initial grant was completed. It is expected that the Branch will be asked to continue through an additional grant.

The RDMQ web page can be accessed through the Branch's web site. The RDMQ User Manual is accessible from a link on this page.

2.4 STRATOSPHERIC OZONE

2.4.1 Arctic Stratospheric Ozone

- H. Fast and R.L. Mittermeier

This annual measurement campaign, to monitor and study the chemistry and dynamics of the arctic stratospheric ozone layer, was conducted from late November 1996 to end of March 1997. Observations were made with lidars, FTIR spectrometers, ozonesondes and a Brewer spectrophotometer.

The measurements and subsequent analyses involved collaboration with the Meteorological Research Institute, the Communications Research Laboratory and the Solar Terrestrial Environment Laboratory in Japan, the University of Denver and Embry-Riddle Aeronautical University in the U.S.A., and the Institute of Space and Terrestrial Science, the University of Saskatchewan and the University of Western Ontario in Canada.

The results show that the polar vortex was late in establishing itself. However, once it did, it remained strong, cold and centered over the north pole much later into spring than in previous years. From the frequent observations of polar stratospheric clouds, as well as elevated levels of ClO (chlorine oxide), it is inferred that chemical and dynamic effects played a major role in decreasing the ozone to the very low values measured this past season.

Persistent strong and cold north polar vortex resulted in very low stratospheric ozone over Arctic

2.4.2 International Ozonesonde "Match" Campaign

- H. Fast and J. Davies

This project optimizes the ozone soundings made at different stations in the northern hemisphere so as to differentiate chemical from dynamic mechanisms of ozone depletion. The campaign started in January and terminated in early April. It involved coordinating ozonesonde launches at the six Canadian stations with those at European stations in collaboration with the Alfred-Wegener Institute for Polar and Marine Research

in Potsdam, Germany. Preliminary results indicated that significant ozone loss had occurred this winter and early spring, especially at higher altitudes and latitudes than in previous winters.

Significant ozone loss was measured, especially at high altitudes and latitudes

2.4.3 Ground-based FTIR Measurements

- H. Fast and R.L. Mittermeier

The purpose of this work is to monitor, simultaneously, a large number of atmospheric gases implicated in stratospheric ozone depletion and the enhanced greenhouse effect, as well as validate measurements carried out by instruments flown on a space shuttle or satellite. The gases can be measured by means of their characteristic spectra. In this past year, solar transmission spectra have been recorded at Egbert, Ontario, approximately twice per month with the new high-resolution Bomem DA8 Fourier transform infrared (FTIR) spectrometer.

Monitoring atmospheric gases with a high resolution FTIR spectrometer system

2.4.4 World Ozone and UV Radiation Data Centre

- E.W. Hare, D.I. Wardle and J.B. Kerr

The World Ozone and Ultraviolet Radiation Data Centre (WOUDC) has two component parts: the World Ozone Data Centre (WODC) and the World Ultraviolet Radiation Data Centre (WUDC).

The WODC processes, archives and publishes world ozone data reported by over 300 stations. These data are published bi-monthly in a publication entitled "Ozone Data for the World" along with an annual catalogue which details all the data received each year. Data are current to the end of 1995. Data output from several data collection and measurement techniques comprise the database input which includes: total column ozone, surface ozone, vertical profile data derived from lidar, ozonesonde and Umkehr techniques. Total ozone measurements are made using several different instruments, most notably the Dobson and Brewer spectrophotometers.

The Brewer also collects high resolution ultraviolet (UV) spectral data and now Japan and Canada regularly submit yearly UV data to be posted on the ftp server for world-wide use. The WUDC has over 110 station-years

of spectral UV data from Canada, the USA, Japan, Argentina and the Antarctic, including the South Pole.

The WOUDC has provided data by means of the Internet (ftp site) since January 1995. International sites routinely submit and extract data by means of the WOUDC ftp site. A new and improved Web site has been developed and may be accessed at www.tor.ec.gc.ca/woudc/woudc.html.

WOUDC released its first CD-ROM of the data archive and distributed to international colleagues

This past year the WOUDC released its first CD-ROM of the data archive and distributed among international colleagues at the Quadrennial Ozone Symposium in Rome, Italy. The WOUDC continues to publish the *Ozone Data for the World* red book which has been a tradition since the early 1960s, however, distribution of the red book has been reduced to semi-annually from bi-monthly.

A joint meeting of the AES and WMO Advisory Committees on Data Management for the WOUDC was hosted by the WOUDC in Toronto in January at which time topics such as UV data exchange, formats and protocols were discussed. This data centre was maintained through the efforts and contributions of E. Carty, V. Fioletov, D. Lynch and K. Parkhill.

2.4.5 ORACLE

- R.M. Hoff and C.T. McElroy

The Branch is participating in the scientific definition of the Ozone Research and Cooperatively Lidar Experiment (ORACLE) satellite and aiding the Canadian Space Agency in defining goals for the use of the data from the mission.

ORACLE would provide capability to monitor ozone in both the stratosphere and the troposphere

ORACLE is one of the first of a new generation of spaceborne instruments, designed to measure the earth's atmosphere. ORACLE consists of an ultraviolet differential absorption lidar (ozone DIAL) placed aboard a polar orbiting satellite. The goal of the ORACLE mission is to obtain vertical profiles of ozone from the surface to the top of the stratosphere with 3 km vertical and approximately 1°x1° (or 100 km x 100 km) horizontal resolution. With this goal achieved, ORACLE would be capable of deriving the most extensive ozone

distribution fields for any planned satellite instrument. These fields are to be used for numerical data assimilation in AES forecast models and would provide unprecedented capability to improve upper tropospheric and lower stratospheric winds and dynamical fields.

ORACLE is a joint program of the CSA and the U. S. National Aeronautics and Space Administration (NASA) and has partnership with AES, Optech Ltd., the Institute for Space and Terrestrial Studies (York University) and Bristol Aerospace.

No other instrument can give ORACLE's capability to obtain vertical profiles of ozone over this range

Optech, Ltd., provided CSA with a definition of the receiver requirements for ORACLE. AES participated in the design review of the instrument and advised CSA on the paths forward toward implementation of the ORACLE design in an upcoming proposal round at both CSA and NASA.

This project is being undertaken in collaboration with E.V. Browell, S. Ismail, S. Crockett and L. McMaster (NASA Langley), A. Dudelzak, F. Rheault and A. Hollinger (CSA), A. Carswell, A. Ulitsky and J. Hahn (Optech), and S. Pal (ISTS).

2.4.6 Space Experiments

- C.T. McElroy

This year, the Space Experiments group again participated in the NASA ER-2 chemistry program by taking part in the summer and winter campaigns of the STRAT (Stratospheric Tracers of Atmospheric Transport) project. The goal was to obtain new information about stratosphere/troposphere exchange and to investigate the rate of exchange of air between low and high latitudes. Two different scientific issues are under study. One is determining the potential effects of proposed stratospheric transport aircraft, and the other is examining in detail the processes by which arctic ozone depletion affects the amount of ozone found at middle latitudes.

Group again participated in the NASA ER-2 chemistry program through STRAT project.

The CPFM (Composition and Photochemical Flux Measurements) experiment was provided for STRAT to make measurements of the direct and scattered solar radiation field in the environment of the aircraft. The

CPFM data are used to calculate the rate at which the ambient radiation field drives some critical photochemical reactions. The radiation field measurements are analyzed using a simple model of the absorption process and estimates of the rate of two important stratospheric reactions are determined. These values are used together with the other data collected on the aircraft to analyze the performance of current photochemical models in explaining the observed composition and dynamics of the upper atmosphere.

The STRAT project involved four measurement campaigns from Ames Research Center near San Francisco and from Barbers Point Naval Air Station in Hawaii. On each deployment, measurements were made spanning the latitude range from just south of the equator to approximately 60 degrees north. Measurements over this broad range of latitudes are required so that the reference composition of the lower atmosphere air which enters the stratosphere at the equator is well known. This is needed so that the correct interpretation of the data collected in the upper atmosphere will be made. The data collected as part of the STRAT project have been released by NASA/Ames as part of their CD-ROM publication, and several papers have been or will be published using this STRAT data set.

The CPMF experiment was originally seen as a simple, direct verification of the radiative transfer codes used to calculate the rates of photochemical reactions in the stratosphere. It has become apparent as the ER-2 chemistry program has advanced that an accurate picture of conditions in the stratosphere can only be constructed and good agreement with the models can only be obtained if two radiation-related and highly variable background parameters are available. These are the surface reflectivity below the flight track and the distribution of ozone above and below the aircraft. The CPMF instrument directly measures the column amount of ozone above the aircraft, and the contribution of radiation scattered from the atmosphere and reflected from the ground below to the total radiation field in the vicinity of the aircraft.

CPFM instrument directly measures column ozone above the aircraft and radiation contributions

In addition to the ER-2 project, the section has continued to support several other experimental activities. These include analyses of Umkehr data to determine the altitude profile of ozone using twilight sky data from the Brewer Ozone Spectrophotometer and advances to UV-Visibility spectroscopy for the measurement of minor constituents from ground-based spectral data. Cooperative research

with York University have provided student support for these activities. Opportunities are also being sought to re-fly the AES SunPhotoSpectrometer which flew first with Astronaut Steve MacLean in 1992.

The Branch cooperated with two foreign laboratories in 1996 to prepare two different proposals to NASA for the construction of satellites under the Earth System Science Pathfinder Announcement of Opportunity. An instrument called OBOE (Ozone Backscatter Observations Experiment) was proposed as part of a submission from the Johns Hopkins University Applied Physics Laboratory (APL), and an instrument named MAST (Measurements of Aerosol in the Stratosphere and Troposphere) was proposed to fly along side the Jet Propulsion Laboratory (JPL) ATMOS infrared spectrometer on a free-flying platform.

The APL experiment was designed to measure ozone, temperature and N₂O profiles at high spatial resolution by combining OBOE data with data from an infrared, nadir-pointing instrument. OBOE is intended to measure ozone using the UV-backscatter technique with a comparable spatial resolution. The JPL experiment is designed to make solar occultation measurements in the UV, visible and infrared parts of the spectrum. The combination of the UV-Visibility and infrared instruments allows the determination of the vertical profile of a large number of stratospheric and tropospheric constituents, as well as the distribution of atmospheric aerosol.

While neither experiment was chosen for funding, they both placed in the top ten of the 40 proposals received with the JPL proposal rated third. The top two proposals were funded and the JPL proposal is considered the official alternate to the two selected in the event that one of them cannot be completed within the assigned budget. As a result, funding at the level of several hundred thousand dollars is being provided to JPL to ensure that the experiment will be ready if it is needed to replace one of the selected proposals.

2.5 CLIMATE CHANGE

2.5.1 Stable Isotope Laboratory

- J.F. Hopper, D. Ernst and A.-L. Norman

The objectives were to develop analytical methodologies for high-precision determinations of stable isotope ratios in atmospheric constituents relevant to atmospheric issues, and to apply these methodologies to analysis of samples relevant to specific research programs, especially climate change.

Several methods for the extraction and determination of isotope ratios (¹³C/¹²C and ¹⁸O/¹⁶O) in samples of

atmospheric carbon dioxide (CO₂) were completed or nearing completion. Procedures for calibrating laboratory working standards to international isotope ratio standards were developed. The large volume method (0.2-2 l of air) produces results with precisions equivalent to those from leading laboratories in the world. Analysis of air in routine flask samples collected at Alert and other sites began in January 1997.

A method for determination of ¹³C/¹²C and ¹⁸O/¹⁶O ratios in samples of atmospheric carbon monoxide (CO) was developed and used at St. Anicet, Quebec, during the NARSTO-CE (North American Research Strategy for Tropospheric Ozone-Canada East) study. Only one previous laboratory and two current laboratories have this analytical capability. Field study data are being used to assess the relative contributions of anthropogenic and non-anthropogenic sources of CO in the area. Preliminary experiments have demonstrated the feasibility of determining ¹³C/¹²C ratios in atmospheric aerosol samples. This is a new research area, with very few previous results reported in the scientific literature due to the need for sensitive modern analytical technology.

Studies demonstrated feasibility of determining ¹³C/¹²C ratios in atmospheric aerosol samples

Components for the introduction and splitting of whole air samples collected in VOC (volatile organic compound) flasks were received. When implemented, these will permit simultaneous quantification of concentrations and ¹³C/¹²C ratios in VOCs. The combined concentration/ isotope ratio data will be used to assess the origins, i.e., biogenic or anthropogenic, of major VOCs in air. The capabilities will also be used to support a collaborative research project with the Institute for Ecotoxicology in Germany which investigates the response of selected VOCs from plant species due to changing atmospheric conditions, such as elevated CO₂ concentrations.

2.5.2 Light Extinction in the Borden Forest

- R.M. Staebler, J.D. Fuentes and M.J. Deary

During the 1995 Borden field study, components of the solar and terrestrial radiation budget were measured both above and below the canopy. The above-canopy instruments were mounted in fixed positions on the tower, whereas the below-canopy measurements were made from a trolley moving on a 30 m track in order to obtain representative data over a statistically meaningful number of tree spacings. In addition, extensive measurements of the leaf area index were made with

plant canopy analyzers, supported by leaf litter collection in the fall.

Data showed a substantial change in the light extinction properties of the canopy during the transitional period of leaf senescence in the month before leaf fall. This period coincides with a significant drop in hydrocarbon emissions by the foliage. These seasonal variations are not presently accounted for in current biogenic emissions models, but will be incorporated in their next generation.

Seasonal changes in canopy characteristics and hydrocarbon emissions examined

In addition, two canopy models relating canopy gap size distributions to canopy architecture have been tested for the first time on a deciduous forest and were found to agree well with our gap fraction and leaf litter data.

2.5.3 Borden Flux Measurements

- R.M. Staebler, H.H. Neumann and M.J. Deary

The exchange of carbon dioxide and energy between the forest at the Borden Forest Research Station and the atmosphere has now been measured continuously since July 1995. The fluxes are measured directly by eddy correlation. Important support parameters, such as radiative fluxes, soil heat flux and moisture as well as a detailed temperature profile, are also monitored. The goal is to continue these measurements over a total of five years to capture a broad spectrum of environmental conditions, and to evaluate the response of the ecophysiological processes that control the fluxes over this spectrum. The program forms part of a network of similar land surface-atmosphere interaction programs across North-America (AmeriFlux) and Europe (EuroFlux). The information gathered by this network is essential in providing constraints to models of the global carbon cycle.

Continuous CO₂ flux measurements at Borden to evaluate impact of climate variability

JOURNAL PUBLICATIONS

Barrett, L. R. McDonald, J. F. ...
Gandy, R. ...
M. ...
Lockhart, D. ...
Smith, G. ...
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