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Edited by Frank Dentener, Terry Keating, and Hajime Akimoto

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Chemical Symbols, Acronyms and Abbreviations

Chemical Abbreviations

C – carbon CH_4 – methane CO – carbon monoxide CO₂ – carbon dioxide F-gases – fluorine-containing halogenated substances that are potent greenhouse gases: hydrofluorocarbons (HFCs), perfluorocarbons (PFCs), and sulfur hexafluoride (SF6) HCHO – formaldehyde Hg – mercurv H_2SO_4 – sulphuric acid HNO₃-nitric acid HO_x – hydrogen oxide radicals Mn – manganese N – nitrogen NH₃ – ammonia N₂O - nitrous oxide N₂O₅ – dinitrogen pentoxide NO - nitric oxide NO₂ – nitrogen dioxide NO₃ – nitrate NO_x – nitrogen oxides NO_v – total inorganic oxidized nitrogen Nr - total reactive nitrogen (including NO_v and NH_x)nss-SO₄ - non-seasalt sulphate OH – hydroxyl radical $O_3 - ozone$ P – phosphorus Pb - lead Rn – radon Si-silicon SO₂ – sulphur dioxide SO₄-sulphate SO_x – sulphur oxides Sr – strontium δ^{34} S – a stable isotope of sulphur

Acronyms and Abbreviations

AAOD – Absorption Aerosol Optical Depth ABC – Atmospheric Brown Cloud ABL – Atmospheric Boundary Layer AC&C – Atmospheric Chemistry and Climate Initiative ACCENT – Atmospheric Composition Change the European Network of Excellence ACE – Aerosol Characterization Experiments ACE-Asia – Asian Pacific Regional Aerosol Characterization Experiment ACS – American Cancer Society ACT – Advanced Combustion Technology AEROCE – Atmosphere/Ocean Chemistry Experiment AEROCOM – Aerosol Comparisons between Observations and Models (a global aerosol model intercomparison project)

AERONET – NASA Aerosol Robotic Network

AERONOX - a soil emissions model

AF – North Africa

AFstY – accumulated stomatal ozone flux

AHSMOG – Adventist Health Study on the Health Effects of Smog

AIM – Asian Pacific Integrated Model

AIRS – Atmospheric Infrared Sounder

ALTO – Airborne Lidar for Tropospheric Ozone

AMS –Aerosol Mass Spectrometer

AOD – Aerosol Optical Depth

AOTxx – Accumulated Ozone Concentration over a Threshold over a Growing Season, where xx is the threshold concentration in ppb

APHENA – Air Pollution and Health: A European and North American Approach

APINA – Air Pollution Information Network for Africa

AQ – Air Quality

AQS – Air Quality Standards

Aqua – A NASA Polar-Orbiting Satellite

AR – Arctic

AS – Asia

ASea – Adjacent Marine Regions

ASIA – Asia excluding the Middle East

ASL – above sea level

- ATLID Atmospheric Lidar, instrument planned for Earth Clouds, Aerosols, and Radiation Explorer satellite mission
- ATS American Thoracic Society

ATSR – Along Track Scanning Radiometer

Aura – a NASA polar orbiting satellite

AVHRR – Advance Very High Resolution Radiometer

BC – Black Carbon

C3 – the photosynthetic mechanism used by most plants

C4 – an alternative photosynthetic pathway

CALIOP – Cloud-Aerosol Lidar with Orthoganol Polarisation

CALIPSO – Cloud-Aerosol Lidar and Infrared Pathfinder Satellite Observations

CAM – Community Atmosphere Model

CAMCHEM– Community Atmosphere Model with Chemistry

CAPMoN – Canadian Air and Precipitation Monitoring Network

CAPTEX - Cross-Appalachian Tracer Experiment

CARIBIC – Civil Aircraft for Regular Investigation of the Atmosphere Based on an Instrument

CASTNET – Clean Air Status and Trends Network

CCB – Cold Conveyor Belt

CCC – Chemical Coordinating Centre

CCN – Cloud Condensation Nuclei

CCSP – U.S. Climate Change Science Program

CDNC – Cloud Droplet Number Concentration

CEIP – Centre on Emission Inventories and Projections

CEMs – Continuous Emission Monitoring Systems

CEREA – Centre d'Enseignement et de Recherche en Environment Atmoshperique

CESY – China Energy Statistics Yearbook

CHASER – Chemical Atmospheric General Circulation Model for Study of Atmospheric Environment and Radiative Forcing CHN – China

CHN-NCP - North China Plain

CHN-NE – NE China

CHN-SE – SE China

CHN-YRB – Yangtze River Basin

CIESIN – Center for International Earth Science Information Network

CIFEX – Cloud Indirect Forcing Experiment

CIRES – Cooperative Institute for Research in Environmental Science

CLE – Current Legislation Scenario

CLRTAP – Convention on Long Range Transboundary Air Pollution

CMAQ – Community Multiscale Air Quality Model

CMAQ DDM - Community Multiscale Air Quality Model with Direct Decoupled Method

CMDL – Climate Modeling and Diagnostics Laboratory

CMIP5 – Climate Model Intercomparison Program #5

CNR – The National Research Council, Italy

CNRS – Centre National de la Recherche Scientifique, France

CONUS – Continental United States

CORINAIR – Coordination of Information on the Environment – Air

COSAM – Comparison of Large Scale Atmospheric Sulphate Aerosol Models

CRF – Concentration-Response Function

CTM - Chemical Transport Model

CUNY – The City University of New York

DA – Dry Airstream

DJF – December, January, February

DLR - Deutsches Zentrum fuer Luft

DMS - Dimethylsulfide

DU – Dobson Units

EA – East Asia

EANET – Acid Deposition Monitoring Network in East Asia

EARLINET – European Aerosol Research Lidar Network

EC – Elemental Carbon

ECB – East Siberia

ECHAM5-HAMMOZ – an atmospheric model (ECHAM), with aerosol (HAM) and tropospheric chemistry (MOZ) modules

ECMWF – European Centre for Medium Range Weather Forecasting

EDGAR – Emissions Database for Global Atmospheric Research

EDGAR-HYDE – An extension of the EDGAR data set back to 1890

EEA – European Environment Agency

EMEP – Cooperative Programme for Monitoring and Evaluation of the Long-range Transmission of Air Pollutants in Europe

I ransmission of Air Pollutants in Europe

ENSO – El Niño Southern Oscillation

ENVISAT – European Space Agency Environmental Satellite

EOTC – European Open Top Chamber

ES – Event Simulations

ETEX – European Tracer Experiment

EU – European Union

FC – Future Climate

FE – Future Emissions

FLEXPART – a Lagrangian particle dispersion model

FO3 – Calculated Stomatal Flux into the Plant

FRSGC – Frontier Research System for Global Change, Japan

Fs – Maximum Forcing at the Surface

FSU – Former Soviet Union

FT – Free Troposphere

GACP – Global Aerosol Climatology Project

- GAINS Greenhouse gas and Air pollution Interactions and Synergies
- GAM Generalized Additive Model
- GASP Aerosol/Smoke Product from the GOES EAST satellite
- GAW Global Atmospheric Watch Programme (within WMO)
- GCAM Global Change Assessment Model
- GCM General Circulation Model
- GEMAQ Global Environmental Multiscale Air Quality Model
- GEMS Global and Regional Earth-system (Atmosphere) Monitoring using Satellite and Insitu Data
- GEO Geostationary Orbit
- GEOS-CHEM A global 3-D atmospheric composition model driven by data from the Goddard Earth Observing System
- GFDL Geophysical Fluid Dynamics Laboratory, NOAA, U.S.
- GFED Global Fire Emissions Database
- GHG Greenhouse Gases
- GICC Gestion et Impacts du Changement Climatique
- GISS Goddard Institute for Space Studies, NASA, U.S.
- GISS-PUCCINI GISS model for Physical Understanding of Composition-Climate Interactions and Impacts
- GMI Global Modeling Initiative
- GOCART Goddard Global Ozone Chemistry Aerosol Radiation Transport
- GOES-EAST Geostationary Operational Environmental Satellite, Eastern Continental United States
- GOME Global Ozone Monitoring Experiment
- GPP Gross Primary Productivity
- GPWv3 Gridded Population of the World, Version 3
- GRACE Gravity Recovery and Climate Experiment
- GRAPE Global Retrieval of ATSR Cloud Parameters and Evaluation
- GWEM Global Wildland Fire Emission Model
- GWP Global Warming Potential
- GWP100 Global Warming Potential Over 100 Years
- HadAM3 U.K. Meteorological Office Hadley Centre Earth Systems Climate Model
- HadGEM U.K. Meteorological Office Hadley Centre Global Environmental Model
- HEI Health Effects Institute
- hPa hectopascal
- HPO Mt. Happo, Japan
- HTAP Hemispheric Transport of Air Pollution
- i Index
- IAGOS Integration of routine Aircraft measurements into a Global Observing System
- IASI Infrared Atmospheric Sounding Interferometer
- ICARTT International Consortium for Atmospheric Research on Transport and Transformation
- ICP International Cooperative Programme
- ICT Intercontinental Transport
- IDC+ Other South East Asia
- IEA International Energy Agency
- IGAC International Global Atmospheric Chemistry
- IGAC Lagrangian 2K4 First Inter-Continental Psuedo-Lagrangian Experiment
- IIASA International Institute for Applied Systems Analysis
- IMAGE -- Integrated Model to Assess the Global Environment
- IMPACT Integrated Massively Parallel Atmospheric Chemical Transport Model
- IMPROVE Interagency Monitoring of Protected Visual Environments
- IN Indian Subcontinent
- INCA Interaction of Chemistry and Aerosol Model
- INDOEX Indian Ocean Experiment

INTEX - Intercontinental Chemical Transport Experiment IPCC – Intergovernmental Panel on Climate Change IPCC AR4 - Intergovernmental Panel on Climate Change Fourth Assessment Report IPCC AR5 - Intergovernmental Panel on Climate Change Fifth Assessment Report IPCC SRES - IPCC Special Report on Emission Scenarios, 2000 IPs - Influence Potentials ITCT - Intercontinental Transport and Chemical Transformation Experiment ITCT-2K2 - Intercontinental Transport and Chemical Transformation 2002 Experiment ITOP - Intercontinental Transport of Ozone and Precursors ITZ – Intertropical Convergence Zone JGCRI – Joint Global Change Research Institute JGSEE - The Joint Graduate School of Energy and Environment JJA – June, July, August JOSIE - Juelich Ozone Sonde Intercomparison Experiment JPN – Japan JPN-E+W – Korea and Mainland Japan JRC - Joint Research Centre K – Kelvin km – kilometers KOR – Korea L – Low LAM - Latin America and Caribbean LATMOS-IPSL – Laboratoire Atmospheres, Milieux et Observations Spatiales-Pierre Simon Laplace Institute LDMz - Laboratoire de Meteorologie Dynamique Zoom model LEO - Low Earth Orbit LLGE - Laboratoire de Glaciologie et Géophysique de l'Environnement LLGHGs - Long-lived Greenhouse Gases LLNL – Lawrence Livermore National Lab, U.S. LMD-IPSL – Laboratoire de Météorologie Dynamique-Pierre Simon Laplace Institute LPDM – Lagrangian Particle Dispersion Models LRT – Long-range Transboundary LRTAP - Long-range Transboundary Air Pollution LS – Lower Stratosphere LWC – Liquid Water Content m – meters M12 – Daylight (12 hours) Growing Season Average M7 – Daylight (7 hours) Growing Season Average MACC - Monitoring Atmospheric Composition and Climate MAF - Middle East plus Africa MAM - March, April, May MARM - Ministry of the Environment, Rural and Marine Media of Spain MBO - Mt. Bachelor Observatory MDA8 O3 - Maximum Daily 8-h Average Ozone ME - Middle East ME00 - 3 Model-mean for 2000s Climate ME90 - 3 Model-mean for 2090s Climate MED – Median MEGAN - Model of Emissions of Gases and Aerosols from Nature MESSAGE - Model for Energy Supply Strategy Alternatives and their General **Environmental Impact** MetOP - a European polar-orbiting meteorological satellite MFR - Maximum Feasible Reduction scenario

mg – milligrams

MILAGRO - Megacity Initiative: Local and Global Research Observations MISR - Multiangle Imaging Spectro-Radiometer MLS - Microwave Limb Sounder MMO1 - Multi-model Mean and Ranges with 2001 Climate MOCAGE - Model of Large Scale Atmospheric Chemistry, Meteo-France MODIS - Moderate Resolution Imaging Spectroradiometer MOPITT – Measurements of Pollution in the Troposphere MOZAIC - Measurement of Ozone on Airbus In-service Aircraft MOZART - Model of Ozone and Related Tracers MOZGN - MOZART modified by GFDL and NCAR MSC-E - Meteorological Synthesizing Centre-East MSC-W - Meteorological Synthesizing Centre-West Mx - Combination of M7 and M12 NA – North America NAAQS - National Ambient Air Quality Standards NAO – North Atlantic Oscillation NAPAP – National Acid Precipitation Assessment Program, U.S. NARE - North Atlantic Regional Experiment NASA – National Aeronautics and Space Administration, U.S. NASA DC-8 – a NASA fixed wing aircraft NATAIR – Natural and biogenic emissions and assessment of impacts on air quality nc - non-crustal NCAR – National Center for Atmospheric Research, U.S. NCLAN - National Crop Loss Assessment Network NCO-P - Nepal Climate Observatory Pyramid NE – Eurasian Boreal NEAQS – New England Air Quality Study NEGTAP - National Expert Group on Transboundary Air Pollution NEI – National Emissions Inventory NEI99 - National Emissions Inventory 1999 ng/m3 – nanograms per cubic meter NH – Northern Hemisphere NIES – National Institute for Environmental Studies (Japan) NILU - Norwegian Institute for Air Research NL – Netherlands NLCS - Netherlands Study on Diet and Cancer NMHCs - Non-Methane Hydrocarbons NMVOCs - Non-Methane Volatile Organic Compounds NOAA - National Oceanic and Atmospheric Administration, U.S. NOAA WP-3D - a NOAA fixed wing aircraft NRC - National Research Council NW - North American Boreal OC - Organic Carbon OECD - Organization for Economic Co-operation and Development OECD90 - Organization for Economic Co- operation Member States as of 1990 OHP - Observatoire de Haute Provence OMI - Ozone Monitoring Instrument PAN – Peroxyacetyl Nitrate PAPA - Public Health and Air Pollution in Asia PATMOS-x - AVHRR Pathfinder Atmospheres Extended project PBL – Planetary Boundary Layer PCFA - Post Cold Front Airstream PFC – Policy Failed Case PgC – petagram carbon

- PHOBEA Photochemical Ozone Budget of the Eastern North Pacific Atmosphere Campaign
- PHOENICS Particles of Human Origin Extinguishing Natural Solar Radiation in Climate Systems
- PHOTOCOMP IPCC Photochemical Model Intercomparison
- PICO-NARE Observatory North Atlantic Regional Experiment Observatory at Mt. Pico, Azores
- PJ Polar Jet
- PM Particulate Matter
- PM2.5 Particulate Matter with a Diameter of 2.5 Micrometers or Less
- PM10 Particulate Matter with a Diameter of 10 Micrometers or Less
- PODy Phytotoxic O3 Dose above a Stomatal Flux Threshold Over a Growing Season
- POET Precursors of Ozone and their Effects in the Troposphere (an EU project)
- POLARCAT Polar study using Aircraft, Remote Sensing, Surface Measurements and Models, of Climate Chemistry, Aerosols and Transport
- POM Polycyclic/Particulate Organic Matter
- POPs Persistent Organic Pollutant
- ppby –Parts per Billion by Volume
- ppty Parts per Trillion by Volume
- PSC Policy Succeed Case
- PSD Class I Class I Areas under the Prevention of Significant Deterioration Rule
- PV Potential Vorticity
- pvu Potential Vorticity Unit
- QRT Quartile
- RAIR Relative Annual Intercontinental Response
- RAPIDC Regional Air Pollution in Developing Countries
- RCP Representative Concentration Pathway
- REAS Regional Emissions Inventory in Asia
- REF Reference Scenario
- RETRO Re-analysis of the tropospheric chemical composition project
- RF Radiative Forcing
- RMT Other Northern Hemispheric Mid-Latitude Regions
- ROSE Rural Oxidants in the Southern Environment study
- RoTAP Review of Transboundary Air Pollution
- ROW Rest of World
- S/R Source-Receptor
- SA Source Attribution
- SA South Asia
- SAEFL Swiss Agency for Environment, Forest, and Landscape
- SCAB South Coast Air Basin
- SCAQS Southern California Air Quality Study
- SCIAMACHY Scanning Imaging Absorption Spectrometer for Atmospheric Chartography/Chemistry
- SCS Six Cities Study
- SE Africa-Asia
- SeaWiFS Sea-viewing Wide Field of View Sensor
- SEI Stockholm Environmental Institute
- SJ Subtropical Jet
- SLC Salt Lake City
- SLCFs Short-Lived Climate Forcers
- SOA Secondary Organic Aerosol
- SON September, October, November
- SOS Southern Oxidant Study
- SP2 Single Particle Soot Photometer
- SPARC Statospheric Processes and Their Role in Climate (part of WMO)

SPRINTARS – Spectral Radiation-Transport Model for Aerosol Species

SRES – IPCC Special Report on Emissions Scenarios

SRx – TF HTAP Source-Receptor Experiment x

SST – Sea Surface Temperatures

STILT - Stochastic Time-Inverted Lagrangian Transport model

STOCHEM – A global three dimensional Lagrangian transport chemical model

STR – Stratosphere

SUM06 – the sum of all hourly ozone concentrations greater than 0.06 parts per million

SW – Low-Latitude Regions of North America

SW DRF - Short-Wave Direct Radiative Forcing

TEMIS – Tropospheric Emission Monitoring Internet Service

TEOM – Tapered Element Oscillating Microbalance

TES – Tropospheric Emission Spectrometer

TexAQS - Texas Air Quality Study

TF HTAP – Task Force on Hemispheric Transport of Air Pollutants

Tg – Teragrams

TM5 – a 3-dimensional atmospheric chemistry-transport zoom model

TOA – Top of the Atmosphere

TOMS – Total Ozone Mapping Spectrometer

TP – TF HTAP Tracer Process Studies

TRACE-A - Transport and Chemical Evolution over the Atlantic

TRACE-P – Transport and Chemical Evolution over the Pacific

TRANSCOM - Atmospheric Tracer Transport Model Intercomparison Project

TROICA-8 – The eighth Trans-Siberian Railway Observations Into the Chemistry of the Atmosphere expedition

 Δ Ts – Global Mean Equilibrium Surface Temperature Response

TSP – Total Suspended Particles

U.S. EPA – United States Environmental Protection Agency

UAVs - Un-manned Aerial Vehicles

UCAR – University Corporation for Atmospheric Research, U.S.

UCI – University of California at Irvine

UJF – Universite Joseph Fourier

U.K. – United Kingdom

UK COMEAP - Committee on the Medical Effects of Air Pollutants, United Kingdom

UK FAAM BAel146 – U.K.'s Facility for Airborne Atmospheric Measurements fixed wing aircraft

ULAQ - an atmospheric chemistry-climate model from the University of L'Aquila, Italy

UM-CAM – a global chemistry-climate model built by the University of Cambridge upon the U.K. Met Office's Unified Model

UNECE - United Nations Economic Commission for Europe

UNFCCC – United Nations Framework Convention on Climate Change

UPMC – University of Pierre et Marie Curie

UPS – Unités Propres de Service (part of CNRS)

USDA ARS – U.S. Department of Agriculture Agricultural Research Service

U.S. – United States of America

UT – Upper Troposphere

UTC – Coordinated Universal Time

UV – Ultraviolet

UVSQ – University of Versailles Saint Quentin

VOCs – Volatile Organic Compounds

W/m2 – Watts per Square Meter

W126 – Weighted Accumulation Over a Growing Season

WCB - Warm Conveyor Belt

WDCGG – World Data Centre for Greenhouse Gases

WHI – Women's Health Initiative observational study

 $\label{eq:WHO} \begin{array}{l} WHO-World \mbox{ Health Organization} \\ WMO-World \mbox{ Meteorological Organization} \\ WRF-Chem-Weather \mbox{ Research Forecast model with Chemistry} \\ WWF \mbox{ G200 eco-regions} - World \mbox{ Wildlife Federation Global 200 eco-regions} \\ ZooM-CiTTy-a \mbox{ high-resolution parcel model with chemistry} \\ \lambda-Climate \mbox{ Sensitivity} \\ \mu g/m3-Micrograms \mbox{ per cubic meter} \\ \mu m-Micrometre \end{array}$

Preface

In December 2004, in recognition of an increasing body of scientific evidence suggesting the potential importance of intercontinental flows of air pollutants, the Convention on Longrange Transboundary Air Pollution (LRTAP Convention) created the Task Force on Hemispheric Transport of Air Pollution (TF HTAP). Under the leadership of the European Union and the United States, the TF HTAP was charged with improving the understanding of the intercontinental transport of air pollutants across the Northern Hemisphere for consideration by the Convention. Parties to the Convention were encouraged to designate experts to participate, and the task force chairs were encouraged to invite relevant experts to participate from countries outside the Convention.

Since its first meeting in June 2005, the TF HTAP has organized a series of projects and collaborative experiments designed to advance the state-of-science related to the intercontinental transport of ozone (O₃), particulate matter (PM), mercury (Hg), and persistent organic pollutants (POPs). It has also held a series of 15 meetings or workshops convened in a variety of locations in North America, Europe, and Asia, which have been attended by more than 700 individual experts from more than 38 countries. The TF HTAP leveraged its resources by coordinating its meetings with those of other task forces and centres under the convention as well as international organisations and initiatives such as the World Meteorological Organization, the United Nations Environment Programme's Chemicals Programme and Regional Centres, the International Geosphere-Biosphere Program-World Climate Research Program's Atmospheric Chemistry and Climate (AC&C) Initiative, and the Global Atmospheric Pollution Forum.

In 2007, drawing upon some of the preliminary results of the work program, the TF HTAP developed a first assessment of the intercontinental transport of ozone and particulate matter to inform the LRTAP Convention's review of the 1999 Gothenburg Protocol (UNECE Air Pollution Series No. 16).

The current 2010 assessment consists of 5 volumes. The first three volumes are technical assessments of the state-of-science with respect to intercontinental transport of ozone and particulate matter (Part A, this volume), mercury (Part B), and persistent organic pollutants (Part C). The fourth volume (Part D) is a synthesis of the main findings and recommendations of Parts A, B, and C organized around a series of policy-relevant questions that were identified at the TF HTAP's first meeting and, with some minor revision along the way, have guided the TF HTAP's work. The fifth volume of the assessment is the TF HTAP Chairs' report to the LRTAP Convention, which serves as an Executive Summary.

The objective of *HTAP 2010* is not limited to informing the LRTAP Convention but, in a wider context, to provide data and information to national governments and international organizations on issues of long-range and intercontinental transport of air pollution and to serve as a basis for future cooperative research and policy action.

HTAP 2010 was made possible by the commitment and voluntary contributions of a large network of experts in academia, government agencies and international organizations. We would like to express our most sincere gratitude to all the contributing experts and in particular to the Editors and Chapter Lead Authors of the assessment, who undertook a coordinating role and guided the assessment to its finalisation.

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André Zuber and Terry Keating

Co-chairs of the Task Force on Hemispheric Transport of Air Pollution

Chapter 2 Observational Evidence and Capabilities Related to Intercontinental Transport of Ozone and Particulate Matter

Lead Authors: Kathy Law, David Parrish

Co-authors: Steve Arnold, Elton Chan, Gao Chen, Owen Cooper, Dick Derwent, David Edwards, Dan Jaffe, Dorothy Koch, Paolo Laj, Randall Martin, John Methven, Paul Monks, Stuart Penkett, Joe Prospero, Patricia Quinn, Lorraine Remer, Johannes Staehelin, Rich Scheffe, Akinori Takami, Hiroshi Tanimoto, Valerie Thouret, Solene Turquety, Christian Zdanowicz, Jerry Ziemke

2.1. Introduction

For several decades it has been possible to measure particulate matter, ozone (O₃) and their important precursors at even the lowest concentrations found in the most remote regions of the Northern Hemisphere. Even the earliest measurements found that long-range transport exerts a strong influence on these observed concentrations. For example, dust of Asian origin was observed throughout the North Pacific region [*Duce et al.*, 1980; *Prospero*, 1979] and studies at the west coast of North America in 1985 identified the influence of Asian emissions on the sulphur budget [*Andreae et al.*, 1988] and on the concentrations of O₃, hydrocarbons, and peroxyacetyl nitrate (PAN) [*Parrish et al.*, 1992]. It has also been clear since at least the 1980s that increasing anthropogenic emissions of NOx since preindustrial times have led to pronounced ozone concentration increases throughout the Northern Hemisphere [e.g., *Crutzen*, 1988].

This chapter aims to document observational evidence for the long-range transport of pollutants between continents. The focus is on the transport of O_3 and particulate matter (PM) from major emission regions of the Northern Hemisphere and their impact on observed concentrations in downwind receptor regions. This includes trans-Pacific and trans-Atlantic transport as well as transport out of Europe and transport to the Arctic. O_3 and a certain fraction of PM such as sulphate are secondary pollutants produced from precursor emissions such as carbon monoxide (CO), NO_x, and sulphur dioxide (SO₂) (see Chapter 1). They are produced close to source regions, especially in the case of PM, before being transported downwind. During transport pollutant concentrations will change due to photochemical production from transported precursors, photochemical and physical loss processes (dry, wet deposition, microphysics) and mixing with air of different composition. Thus, there is no point-to-point relationship between source and receptor. Air masses arriving at a downwind location will be the result of complex air mass histories, and will include components related to emissions from various source regions.

The signature of long-range pollutant transport in downwind measurements depends on the lifetime of the particular pollutant and the extent to which a pollutant plume mixes with other air masses. For PM and other pollutants with short lifetimes, on the order of a few days, long-range transport between continents is observed in the form of discrete plumes with pollutant concentrations significantly greater than those usually encountered. Often such a plume can be directly attributed to a particular upwind source. In favourable cases particular plumes can be tracked in satellite data over periods of several days.

For O₃ and CO, pollutants with lifetimes of weeks to months in the free troposphere, mixing of air masses with different histories plays a dominant role in defining the spectrum of concentrations found in any region of the troposphere. Photochemical production can also be important for O₃. Mixing can significantly modify discrete plume signatures. Figure 2.1 shows the concentration distributions of CO and O₃ derived from 176 separate ascents and descents of MOZAIC aircraft into two North American west coast cities during the summers of 2003-2006. These distributions exemplify the plume signatures that result after trans-Pacific transport. At each city the CO distribution is well defined as a log-normal distribution of mixing ratios, with only a very few ($\approx 1 - 2$ % of the total data) higher concentration plumes distinguishable from this general distribution, and only at Portland, Oregon (U.S.). Similarly, the O₃ distributions exhibit very few plumes (<1 % of the data) with concentrations greater than 100 parts per billion by volume (ppbv), which generally reflect air masses with a stratospheric origin. Thus, the entire troposphere can be envisioned as completely filled with plumes of continuously varying concentrations in the process of intermixing and dispersing. The broad distribution of concentrations represents the plumes responsible for transporting the large majority of O_3 and CO between continents, and the discrete, more concentrated plumes contribute only minimally. Impacts from a particular emission region will therefore be reflected in the average and associated variability of observed concentrations at a particular downwind location, and not as a series of discrete events. The fact that air arrives over a downwind continent as a continuous distribution of air masses with different emission histories from both anthropogenic and natural sources makes it difficult to quantify the contribution directly attributable to a particular emission region from observations alone.

Abundant evidence of episodes of long-range transport of elevated concentrations of both O₃ and PM is available as exemplified in this chapter, but determination of the impact of observed transport on downwind regional air quality presents further challenges, i.e. how does import of pollutants from a particular source region change concentrations in the boundary layer of the receptor region. It is clear there are preferential regions for pollutant import over receptor regions (see Chapter 1), but import of pollutants in the free troposphere does not mean that these air masses then descend into the boundary layer. Robust, quantitative determination of the pollution import into a region would require a dense network of vertically resolved measurements, which presently does not exist.

Various definitions have been employed in attempts to quantify the contribution of long-range transport to the observed concentrations and temporal changes in concentrations of pollutants observed in downwind measurements (see Chapter 1). The term —backgound" is often used to describe concentrations at —clea, remote" sites that have not been influenced by anthropogenic pollution. However, for relatively long-lived pollutants such as O₃, all sites in the northern hemisphere are influenced by anthropogenic emissions, so that the use of this term is ambiguous. The term "baseline" is used here to describe concentrations in air masses without the contribution from local anthropogenic emissions. Diagnosis of baseline concentrations is not straightforward because of the complex air mass histories described above. Analysis of decadal changes in mean O₃ concentrations at representative receptor region sites can provide indications of the impact of changing anthropogenic emissions. However, it must be recognized that changes in the observed concentrations can be confounded by changes in natural sources and changes in transport patterns. Furthermore, different changes of anthropogenic emissions in different source regions can complicate the picture. Thus, observing concentration changes and attributing them to changes in upwind emissions is complex.

In this Chapter, we first consider the direct observational evidence for long-range transport of O₃ (Section 2.2) and PM (Section 2.3) from satellite, aircraft, and ground-based data. Long-term changes (trends) in the amount of O₃ or PM at appropriate measurement locations are also discussed. Section 2.4 summarizes results arising from the use of meteorologically-based and data-based techniques (e.g. constituent ratios) to quantify the sources contributing to measurements collected at particular locations. Results and subsequent analysis from field experiments specifically designed to quantify the processing occurring during long-range transport are also discussed. A summary of future research needs is given in Section 2.5. In this chapter the term —arosols" is often used interchangeably with —paiculate matter (PM)" as is customary in the field of atmospheric chemistry, even though —arosols" technically refers to the suspension of PM in air.

FINDING: The signature of long-range pollutant transport in measurements made downwind of sources depends on the lifetime of the particular pollutant. Discrete plumes of enhanced concentrations characterize pollutants with short lifetimes and no photochemical sources, while continuous distributions of concentrations represent transport of pollutants with longer lifetimes. In the latter case, the entire troposphere can be envisioned as completely filled with plumes of continuously varying concentrations in the process of intermixing and dispersing.

RECOMMENDATION: For longer-lived species such as O₃ and CO a broad distribution of concentrations represents plumes responsible for transporting the large majority of these species between continents. This broad distribution of baseline concentrations of such species

requires fuller characterization by measurement programs to allow comparison with global model calculations, and for input as boundary conditions into regional air quality models.



Figure 2.1. Summertime probability distribution functions of CO (red lines) and O_3 (blue lines) measured between 2 and 10 km altitude by MOZAIC (http://mozaic.aero.obs-mip.fr) aircraft on descents into and ascents out off Portland, Oregon (solid lines) and Los Angeles, California (dotted lines) on the U.S. west coast.

2.2. Long-range Transport of Ozone and its Precursors

2.2.1. The View from Satellites

The major advances in satellite remote sensing over the last decade now provide a global perspective of intercontinental pollution transport and the impact of that transport upon downwind receptor regions. Table 2.1 summarizes satellite remote sensing capabilities of tropospheric O₃ and its precursors. Only downward (nadir) viewing instruments are included here. All fly in near-polar, sun-synchronous, low Earth orbits. Observations at low and mid-latitudes are at a constant local time; sampling frequency increases at high latitudes. These instruments employ passive techniques, observing either solar backscatter (SCIAMACHY, OMI, GOME-2) or thermal emission (MOPITT, TES, AIRS, IASI). Retrievals of NO₂ and formaldehyde (HCHO) are tropospheric columns, while the retrievals of CO and O₃ often have some vertical profile information (typically 0.5-2 Degrees of Freedom for Signal). Other observations include nitric acid (HNO₃) [*Wespes et al.*, 2009] and glyoxal [*Wittrock et al.*, 2006]. Two recent workshops assessed air quality applications from space [*Edwards*, 2006; *EU*, 2006], and two recent reviews describe capabilities for satellite remote sensing of air quality [*Fishman et al.*, 2008; *Martin*, 2008].

Satellite observations of CO provide the most compelling remote-sensing evidence for longrange transport of trace gases. Animations of satellite observations for successive days reveal clear visualization of transport from continental source regions across oceans. Published examples of longrange transport of CO within the Northern Hemisphere include transport both from Asia to North America [*Heald et al.*, 2003; *Zhang et al.*, 2008] and from North America to Europe [*Guerova et al.*, 2006; *Pfister et al.*, 2006]. Figure 2.2 shows a multi-day average of a CO plume from intense wildfires in Alaska and Canada. The plume can be traced across North America and the Atlantic Ocean to Europe. Aircraft measurements as part of the ICARTT aircraft campaign that sampled this plume corroborate the long-range transport. Inverse modelling using the MOZART chemical transport model showed that the fires emitted about as much CO as did human-related activities in the continental U.S, during the same time period, about 30 teragrams (Tg) CO for June-August, 2004. Modelling and measurements show that emissions from the 2004 North American wildfires caused ground-level concentrations of O₃ to increase by 25 per cent or more in parts of the northern continental U.S. and the central North Atlantic and by 10 per cent as far away as Europe [*Pfister et al.*, 2006; *Val Martin et al.*, 2006, see also discussion in Section 2.4]. Satellite observations of NO₂ also provide evidence of long-range transport from North America to Europe [*Guerova et al.*, 2006; *Martin et al.*, 2006].

Satellite observations of tropospheric O_3 columns have matured considerably over the last decade to reveal mid-latitude features with increasing confidence [*Creilson et al.*, 2003; *Liu et al.*, 2010; *Schoeberl et al.*, 2007; *Worden*, 2007; *Ziemke et al.*, 2006]. Figure 2.3 presents examples for a summer and an autumn month (northern hemisphere). The O_3 columns shown are dominated by free-tropospheric concentrations, and elevated topography reduces local O_3 columns (i.e. western North America, Himalayas). Nonetheless, significant O_3 enhancements are apparent in regions of large precursor emission, for example, the eastern U.S. [see also *Creilson et al.*, 2003] and East Asia in July. A broad enhanced region in the northern mid-latitudes is associated with export of O_3 and its precursors from adjacent continental regions [*Ziemke et al.*, 2006]. The enhancement over the Middle East reflects a combination of lightning, pollution sources, and dynamics, as inferred from GEOS-Chem simulations [*Liu et al.*, 2009]. Lightning, biomass burning, and stratospheric O_3 contribute to the O_3 column enhancements in the southern tropics in October, as indicated by GEOS-Chem simulations [*Sauvage et al.*, 2007].

Table 2	.1.	Current	nadir	-viewing	satellite	remote	sensing	of troposj	oheric	ozone and	l its	precursors
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Instrument	Platform	Meas.	Typical Nadir Res.	Equator Crossing	Global coverage	Spectral Range	NO ₂	НСНО	СО	O ₃
		Period	(km)	Time ^a	(days) ^b	(µm)				
MOPITT	Terra	2000-	22x22	10:30	3.5	2.3, 4.7			Х	
AIRS	Aqua	2002-	14x14	1:30	1	3.7-16			Х	
SCIA-	Envisat	2002-	60x30	10:00	6	0.23-2.3	Х	Х	Х	Х
MACHY										
OMI	Aura	2004-	24x13	1:45	1	0.27-0.50	Х	Х		Х
TES	Aura	2004-	8x5	1:45	n/a	3.3-15.4			Х	Х
GOME-2	MetOp	2006-	80x40	9:30	1	0.24-0.79	Х	Х		Х
IASI	MetOp	2006-	12x12	9:30	0.5	3.6-15.5			Х	Х

^aCrossing time occurs at both AM and PM.

^bValue given for clear-sky conditions. Clouds impede the retrieval.

^cMOPITT: Measurements Of Pollution In The Troposphere; AIRS: Atmospheric Infrared Sounder;

SCIAMACHY: SCanning Imaging Absorption SpectroMeter for Atmospheric CHartographY; OMI: Ozone Monitoring Instrument; TES: Tropospheric Emission Spectrometer; GOME: Global Ozone Monitoring Experiment; IASI: Infrared Atmospheric Sounding Interferometer

2.2.2. Direct Evidence for O₃ and Precursor Transport from In Situ and Lidar Measurements

Because of the complexity of its sources, correlations of in-situ observations of O_3 with other tracers is the most certain and direct means to identify episodic long-range transport of O_3 of anthropogenic origin. Tracers such as CO, NO_x, non-methane hydrocarbons, mercury (Hg) and aerosols have all been used to help identify sources of O_3 in polluted airmasses. The quantitative relationship between O_3 and a tracer is often expressed as an enhancement ratio, which is the ratio of the increase of O_3 to that of the tracer during the transport episode (see Section 2.4.2). Numerous studies have identified long-range transport of anthropogenically generated O_3 across the Pacific and/or Atlantic Oceans. In some cases, intercontinental transport of O_3 has been shown to significantly impact the surface concentrations and even contribute to exceedances of local ambient air quality standards (see Section 2.2.4).

Trans-Atlantic transport

Many studies have documented long-range transport of O₃ and its precursors across the North Atlantic [*Honrath et al.*, 2004; *Huntrieser et al.*, 2005; *Stohl and Trickl*, 1999; *Stohl et al.*, 2003; *Trickl et al.*, 2003; *Val Martin et al.*, 2006]. Sources include both boreal fire plumes as well as urban and industrial pollution. The transport occurs mostly in the free troposphere and has been detected by aircraft and lidar





Figure 2.2. Example of long-range transport from Alaskan and Canadian wildfires to Europe observed by MOPITT over 15-23 July 2004. Plumes of anthropogenic pollution can also be seen leaving Asia and crossing the Pacific Ocean. [Reprinted with permission from Figure 2.6 in *Global Sources of Local Pollution: an Assessment of Long-Range Transport of Key Air Pollutants to and from the United States*, [2010], by the National Academy of Sciences, Courtesy of the National Academies Press, Washington, D.C.]



Figure 2.3. Example of tropospheric ozone columns determined by residual from OMI observations of total column O₃, and MLS observations of stratospheric O₃ [*Ziemke et al.*, 2006]. Values reflect a complex interplay of long-range transport, in situ chemical production, loss processes, and stratospheric sources.

and at surface sites in the Alps. At Mace Head, a low elevation site on the west coast of Ireland, Derwent et al. [1997] found 5 probable cases of North American influence but the concentrations were quite low. The only major North American export event to show a strong direct impact on low altitude European sites involved smoke plumes from the widespread forest fires in Canada [*Forster et al.*, 2001].

The International Global Atmospheric Chemistry (IGAC)-sponsored ITCT Lagrangian 2K4 experiment, which was part of the ICARTT, ITOP and INTEX-A field campaigns in summer 2004, provided the opportunity to observe (and model) in situ formation of O₃ during transport from emission regions in North America. In total 4 aircraft were used, along with surface and ozonesonde observations [*Fehsenfeld et al.*, 2006]. Measurements of CO, black carbon (BC), NO_x, total reactive nitrogen (NO_y) and O₃ were also made at the PICO-NARE Observatory (38.5°N, 28.4°W) at 2.2 kilometres above sea level (km asl) in the Azores. This site is well situated to observe long-range transport since the Azores are frequently impacted by airflow from middle and high latitudes [*Honrath et al.*, 2004; *Owen et al.*, 2006; *Val Martin et al.*, 2008]. Indeed, North American emissions emitted 6 to 15 days earlier, frequently impacted the PICO-NARE station during summer 2004 [*Val Martin et al.*, 2006]. Figure 2.4 shows an example that included the highest concentration of CO yet recorded at the station.



Figure 2.4. Time series of 30-min average CO, NO_y and O_3 , and 1-hour average BC observations at the PICO-NARE Observatory (Azores, Portugal) during transport of boreal wildfire emissions to the site. CO is plotted with open blue circles, NO_y with red squares, BC with open purple triangles and O_3 with green triangles. Vertical solid lines bound the transport event. [Adapted from Figure 3 in Val Martin, M., et al. (2006), Significant enhancements of nitrogen oxides, black carbon, and ozone in the North Atlantic lower free troposphere resulting from North American boreal wildfires, *Journal of Geophysical Research*, 111(D23S60).]

In summer 2004 two ground-based ozone lidars at Observatoire de Haute Provence (OHP) in south France (43.9°N, 5.7°E, 0.7 km asl) measured vertical profiles of O_3 and aerosol scattering ratio from the boundary layer to the tropopause (Figure 2.5). The largest variability in the columns was due to local pollution within the boundary layer and stratosphere-troposphere exchange. Several O_3 rich layers within the free troposphere also had aerosol enhancements and were related to long-range transport of biomass burning emissions from North America [*Real et al.*, 2007]. These polluted layers were thin (< 1 km) and remained coherent as they were transported over the Atlantic Ocean. The layers observed above OHP exhibited O_3 mixing ratios 50% greater than values in adjacent air and increased the tropospheric column O_3 by 5 to 10% [*Ravetta et al.*, 2007]. In 2004 aircraft as part of the MOZAIC program also measured fire plumes originating from Alaska in the free troposphere over the eastern U.S., the North Atlantic and Western Europe [http://mozaic.aero.obs-mip.fr, *Elguindi et al.*, 2010]. Note that while CO was strongly enhanced in the MOZAIC vertical profiles, O_3 was not. Detailed analyses of such cases and in particular Lagrangian cases where the same air masses were sampled several times have shown significant O_3 production in certain plumes (e.g. forest fire plumes) whilst in other cases mixing and photochemical destruction dominated. Further examples are discussed in Section 2.4.



Figure 2.5. (a) Ozone mixing ratio cross section in ppb measured at Observatoire de Haute Provence in July 2004. Tick marks correspond to 0000 UT. This figure combines vertical profiles measured by 2 ozone lidars. ECMWF analyses have been used to compute potential vorticity (PV) profiles in order to estimate the height of the tropopause (PV = 1 pvu for the lower thick black line, PV = 2 pvu for the upper one) and spot stratospheric intrusions (large squares). (b) Vertical cross section of aerosol scattering ratio (a quantity proportional to the aerosol concentration) derived from the 316-nm wavelength ALTO lidar on the same days. Ozone rich layers related to long-range transport are circled in black. [Adapted from Ravetta, F., et al. (2007), Long-range transport and tropospheric ozone variability in the western Mediterranean region during the Intercontinental Transport of Ozone and Precursors (ITOP-2004) campaign, *Journal of Geophysical Research*, *112*(D10S46).]

Trans-Pacific transport

The greater transport distance and increased complexity of transport patterns make it more difficult to detect long-range transport across the Pacific Ocean. Our understanding of trans-Pacific transport of O₃ and other pollutants has been improved by recent studies, e.g. the NASA led INTEX-A and B campaigns, which measured from aircraft over North America and the eastern North Pacific [Singh et al., 2006; Singh et al., 2009] and from the Mt. Bachelor Observatory (MBO) at 2.7 km asl in central Oregon [Jaffe et al., 2005a; Weiss-Penzias et al., 2007]. During INTEX-A, which took place in the summer of 2004, several plumes of Asian origin were encountered as far downwind as over eastern North America [e.g., Liang et al., 2007]. Fischer et al. [2009] report on PAN decomposition and O₃ production during subsidence based upon observations from the Mt Bachelor Observatory. This work indicates that transport of PAN and its subsequent decomposition to NO_x has a significant impact on the O₃ concentrations in air arriving at North America. Reidmiller et al. [2009] report on interannual variability in long-range transport using satellite data and observations from Mt. Bachelor, combined with the GEOS-CHEM model. They conclude that variations in transport and biomass burning emissions largely drive the year-to-year variations in CO seen in the eastern Pacific atmosphere. Weiss-Penzias et al. [2007] identified ten separate episodic pollution transport events from Asian industrial sources arriving at Mt. Bachelor. In these events the Hg/CO enhancement ratio appears to be a unique tracer of Asian pollution plumes, being 3-4 times higher in Asian plumes compared to U.S. or biomass burning plumes.

In the past few years efforts have been made to measure pollutant concentrations in air masses above East Asia, especially within China where emissions are rapidly increasing. These studies [*Ding et al.*, 2009] have revealed pollutant concentrations that are much greater than those encountered over Europe or North America [e.g. *Hudman et al.*, 2007; 2008]. High pollutant levels detected above the boundary layer over China include a plume at 2.6 km above northeast China with O₃ and CO as high as 140 ppbv and 1185 ppbv, respectively [*Ding et al.*, 2009] and a layer of pollution at 2.5-3.5 km above Beijing, lofted by upslope mountain flow, with 90 ppbv of O₃ and 2200 ppbv of CO. Ding et al. [2008] examined O₃ from the MOZAIC campaign and found higher concentrations in the lower troposphere then other large Northern Hemispheric cities. These strong Asian pollution plumes contain far more CO than has been detected in Asian plumes transported to above western North America [maximum CO of ~300 ppbv, *Nowak et al.*, 2004], indicating the degree of dilution that occurs during transport.

Transport across South Asia, Europe and East Asia

The impact of European pollution on Asia has been demonstrated from ground-based observations at Mondy in East Siberia [*Pochanart et al.*, 2003]. During the TROICA-8 expedition [*Turnbull et al.*, 2009] Δ^{14} CO₂ was measured together with CO, sulphur hexafluoride and perchloroethylene along the Trans-Siberian railway between Moscow (Russia, 55°44' N, 37°33' E) and Khabarovsk (Russia, 48°33' N, 135°06' E) in March-April 2004. They observed an increase of Δ^{14} CO₂ (a measure of the fossil fuel contribution to measured carbon dioxide concentration) and a decrease of CO from West to East, which they interpreted to reflect emission and transport of anthropogenic species from Europe to Asia. More recently, the Nepal Climate Observatory-Pyramid (NCO-P) at 5.1 km asl was established in the Khumbu valley in the Himalayas [*Bonasoni et al.*, 2008]. Figure 2.6 shows O₃ and BC measured at the observatory. Pollutant transport with high O₃ (63±9 ppbv) to the site is common in the pre-monsoon period. According to back-trajectory analysis, they concluded that it was probably related to continental outflow from Eurasia with an upper troposphere/lower stratosphere influence. O₃ enhancements occurred both with and without enhancements in other tracers. In mid-June O₃ and BC were transported to the site from anthropogenic sources to the east near the India-Pakistan border.



Figure 2.6. O₃ and BC measured March-August 2006 at the Nepal Climate Observatory-Pyramid (NCO-P) at 5.1 km asl in the Khumbu valley of the Himalayas. [Adapted from Figures 6 and 8 from Bonasoni, P., et al. (2008), The ABC-Pyramid Atmospheric Observatory in Himalaya for aerosol, ozone and halocarbon measurements, *Science of the Total Environment, 391*: 252-261, with permission from Elsevier.] O₃ is shown in red circles on left axis (ppbv) and BC in blue crosses on right axis (nanograms per cubic meter). The first high O₃ event in early March was attributed to UT/LS transport due to meteorological conditions and an absence of other pollutant tracers. The high O₃ event in mid-June, which also showed a BC enhancement, was attributed to anthropogenic pollution arriving from the east.

2.2.3. Indirect evidence from long-term trends in baseline O_3

During the latter half of the 20^{th} century baseline concentrations of O₃ increased markedly at northern mid-latitudes. This increase has been documented by a variety of observational studies, and is generally attributed to increasing anthropogenic emissions of the precursors that fuel photochemical O₃ production. These increased emissions accompanied the growth and development of the economies of the industrialized nations [e.g., *Horowitz*, 2006; *Lamarque et al.*, 2005]. This observed increase in O₃ concentrations is one of the most important manifestations of the hemispheric transport of air pollution. Uncertainty remains regarding the magnitude of the increase from pre-industrial to present

day since pre-industrial O_3 concentrations are poorly known, few measurements were made before the late 1970s, and different data sets for the same region do not always give consistent results. Since global models cannot reproduce the very low O_3 levels suggested by the measurements of that time [e.g. *Mickley et al.*, 2001], the quantitative assignment of the cause of the increase is also uncertain. This section reviews the strongest observational evidence for these temporal trends. The primary focus is on the O_3 concentrations in onshore flow at the west coasts of Europe and North America with some comparisons to trends observed in Asian outflow. Without a west coast, it is more difficult to establish long-term trends for baseline O_3 concentrations in onshore flow most clearly show the influence of long-range transport, since they represent baseline O_3 concentrations.

Figures 2.7 and 2.8 summarize the results from several data sets that provide the strongest evidence for increasing baseline O₃ concentrations at northern mid-latitudes since the mid-20th century. Figure 2.7 shows results from spring. In this season intercontinental flow is most important, observational data are most abundant, and influences from regional photochemical O₃ production are relatively small. Figure 2.8 compares springtime trends with those in other seasons. These data represent the longest, highest quality measurement records available from sites that are likely to represent baseline O₃ changes. The studies vary in the time period covered, and in the degree to which regional influences may obscure baseline trends. Given this variability, a simple statistical approach is chosen. Linear and quadratic polynomial least-squares fits are made to seasonally averaged data from each site. Baseline data are examined where available (i.e. only at Mace Head and Pacific marine boundary layer). The polynomial fit is included in Figure 2.7 only if the quadratic term is significant. At least up to the year 2000 the linear trends have remained remarkably constant over many of the individual data records. One can argue that there has been a levelling off or even decrease at the European stations, particularly at the three central European sites (Hohenpeissenberg, Jungfraujoch and Zugspitze) where the quadratic term is statistically significant, and negative. For these three sites the average trends in Figure 2.8 are derived from the linear-least squares fits made to the portion of the data record before 2000. For all sites the slope of the linear trend derived from this statistical approach is the best estimate of the average annual change in the measured O_3 for the season and period selected. In all cases 95% confidence limits are calculated for the slope. Unfortunately, very few data sets have been selected for baseline conditions, but all the sites are located where trends in baseline O₃ are expected to dominate the trends in average O₃. Consistency in the derived trends over large regions supports the assumption that the derived trends represent the baseline O₃ trend.

Beginning in the 19th century a long record of apparently reliable O₃ measurements in ambient air was collected in Europe. Measurements performed at the Montsouris Observatory close to Paris from 1876 to 1912 using a solution chemistry method show low O_3 concentrations (10-15 ppby) in the planetary boundary layer (PBL) [Volz and Kley, 1988]. Similar measurements were conducted from 1956-1983 at Arkona, a coastal site on the Baltic Sea [Feister and Warmbt, 1987], and the series was continued at the nearby Zingst site (Figures 2.7 and 2.8). The earliest springtime measurements at Arkona show O₃ concentrations around 15 ppbv followed by an increase (amounting to about 20 ppbv) to the present Zingst concentrations. However, it remains difficult to judge whether SO₂, an interference of the chemical method, produced a negative bias in these earlier measurements, although data affected by SO₂ transport from the capital of Paris were removed by using local wind measurements at the Montsouris Observatory [Volz and Kley, 1988]. Solution chemistry measurements performed at Arosa, an alpine valley site at 1.8 km asl in the Swiss Alps in the 1950s also showed low annual mean values of around 20 ppbv [Staehelin et al., 1994], and O₃ was also measured in Arosa during some clear nights in the 1930s (using long-path ultraviolet measurements). These 1930s measurements yielded similar values to the 1950s solution chemistry measurements, which supports their reliability (note that the large increase in anthropogenic O_3 precursor emissions took place in the decades after World War II).



Figure 2.7. Springtime trends in O_3 concentrations measured in (a) Europe and (b) western North America and Japan. The lines (in colour) indicate the linear regressions to the data, and the curves (in black) indicate quadratic polynomial fits to the three central European sites over the time span of the lines. Arkona and Zingst are two sites located close to the Baltic Sea. Mace Head is located at the west coast of Ireland. Hohenpiessenberg (1.0 km asl) and Zugspitze (3.0 km asl) are in southern Germany, and Jungfraujoch (3.6 km asl) is in Switzerland. The North American data are from several sea level Pacific coastal sites and Lassen National Park (1.8 km asl) near the west coast, and from the free troposphere over the western part of the continent. The Japanese data are from Mt. Happo (1.9 km asl) on the Japanese mainland and Rishiri, a northern (45N) sea level island site.

Overall, a consistent trend of increasing O₃ has been observed at European surface sites through the second half of the 20th century, beginning with the early measurements summarized above, and continuing with modern measurement methods. Figure 2.7a shows that long, continuous European surface O₃ records from the rural Hohenpeissenberg (Stefan Gilge, Hohenpeissenberg Meteorological Observatory, German Meteorological Service, Hohenpeissenberg, Germany, personal communication, 2010) and the mountaintop Zugspitze (Hans-Eckhart Scheel Karlsruhe Institute of Technology, IMK-IFU, personal communication, 2010) sites in southern Germany, as well as the shorter records for the marine Mace Head [*Derwent et al.*, 2007] and the mountaintop Jungfraujoch sites [*Bronnimann et al.*, 2002], all approximately parallel the Arkona-Zingst springtime record, at least until 2000. A similar trend is also seen in continuous surface O₃ measurements at another alpine high altitude site: Sonnblick (Austria, 3.1 km asl). The data from the Figure 2.8 demonstrate that O₃ has increased during all seasons with some indication of greater increases in spring and winter, and
smaller increases in summer and particularly autumn. Trends determined from springtime data sets from Arosa in the 1930s, 1950s and around 1990 [*Staehelin et al.*, 1994] and summertime data from Jungfraujoch in the 1930s [*Crutzen*, 1988], which define the end points of the trends for those two sites in Figure 2.8, are consistent with the trends extracted from the continuous data sets.



Figure 2.8. Average annual linear trends in O_3 concentrations in different seasons in Europe (left of dashed lines), North America and Japan (right of dashed lines). The slopes of linear regressions of seasonally averaged data (e.g., the lines in Figure 2.7) provide the trends. The sites, symbols and period of the linear trend determination are the same as in Figure 2.7, except for the three central European sites, where the linear fits end in 2000.

Trends derived from the more limited North American measurements (Figures 2.7 and 2.8) are consistent with each other and similar to the European trends although they continue after 2000. No long-term record is available from a single North American marine boundary layer site that is suitable for characterizing baseline O_3 concentrations arriving at North America. However, Parrish et al. [2009] combined baseline data from several North American marine sits to derive seasonal baseline trends (included in Figures 2.7 and 2.8) that are approximately the same as those from Mace Head. Similarly, trends at a long-term, rural site near the west coast of North America that can be considered to represent trends in O_3 in onshore flow (Lassen National Park) parallel the marine boundary layer trends. A recent study that combined all available free troposphere data for springtime over western North America [*Cooper*, 2010] derived a trend in approximate accord with the trends derived from the surface measurements, and the trends were stronger in air with strong influence from the South and East Asian boundary layer. Although shorter than the European records, the three North American trends indicate increasing baseline O_3 in air entering North America from at least the mid-1980s to the present.

At inland North American sites consistent baseline O_3 concentration trends are difficult to discern. Jaffe and Ray [2007] report that average O_3 concentrations generally increased from 1987– 2004 at rural sites in the western US, but the rate of increase varied from 0 to 0.5 ppbv/yr across 9 sites. Oltmans et al. [2006] find no generally increasing trends in average O_3 concentrations over North America. It may well be that at interior North American locations, the increase in baseline O_3 concentrations is obscured by decreasing regional and local O_3 production due to decreasing emissions of O_3 precursors within North America. Indeed, the US Environmental Protection Agency (http://www.epa.gov/airtrends/ozone.html#oznat) notes that maximum O_3 concentrations are decreasing throughout the country in response to reduced precursor emissions. Long-term, continuous O_3 measurements are limited in Asia, but those that exist generally indicate increasing O_3 concentrations. In Japan, continuous measurements at Mt. Happo Observatory (1.9 km asl) show a springtime O_3 increase of 1.30 ± 0.28 ppbv/yr from 1991 to 2007 [*Tanimoto*, 2009] and positive trends in all seasons that are the most rapid increases found in any of the data sets investigated here (see Figure 2.7b and 2.8). Six Pacific Rim island sites [*Tanimoto et al.*, 2009], e.g. Rishiri Island included in Figure 2.7, together give on average an increasing springtime trend of 0.62 \pm 0.36 ppbv/yr, but trends from individual stations are not statistically significant due to their short time span. West of Japan, Beijing experienced annual average O_3 increases in the PBL of 5-8 ppbv between 1995 and 2005, as measured by MOZAIC aircraft profiles [*Ding et al.*, 2008].

Increasing export of O₃ from Asia is expected from observed increases in regional O₃ concentrations. In the region of Taipei, Taiwan (25° N), yearly surface O₃ averaged across several sites increased during 1994-2003 at the rate of 0.96 ppbv/yr [*Chou et al.*, 2006]. In southern China, average yearly O₃ increased by about 50% between the late 1980s and 1990s at an urban monitoring site in Hong Kong (22° N) [*Chan et al.*, 2003]. A coastal site southeast of Hong Kong shows an O₃ rate of increase of 0.58 ppbv year⁻¹ for yearly data during 1994-2007 [*Wang et al.*, 2009]. In East Asia, generally all sites show that O₃ has increased over the past 20 years as would be expected from the strong increases in regional O₃ precursor emissions (e.g. see Figs. 3.9 and 3.10). The rate of increase is on the order of 1 ppbv/yr, somewhat stronger than the rate of increase in Europe and North America.

Ozonesondes provide some of the longest-term records of the evolution of baseline O₃ over Europe, as well as North America and Japan, but their usefulness appears limited. The three long-term European records (based on Brewer Mast sensors at Hohenpeissenberg, Germany; Payerne, Switzerland; and Uccle, Belgium) provide the longest continuous records of O_3 in the free troposphere. They show strongly increasing concentrations from 1970 to 1990, but the three records show large deviations [see Logan et al., 1999]. The O₃ evolution derived from the ozonesonde measurements of Hohenpeissenberg and Payerne do not agree well with the nearby mountain surface series of Jungfraujoch and Zugspitze in the early part of the 1990s [Ordóñez, 2006]. The O₃ changes obtained from regular aircraft measurements of MOZAIC data since 1994 [Thouret et al., 2006; Zbinden et al., 2006] are more consistent with the surface O_3 measurements of Jungfraujoch and Zugspitze than those of the ozonesonde records from Hohenpeisseneberg and Payerne. A recent comparison of O_3 changes in the upper troposphere derived from climatologies of the regular aircraft measurements from the project GASP (1975-1979) and MOZAIC (1994-2001) programs showed unexplained differences with the changes derived from the ozonesonde measurements [Schnadt *Poberaj et al.*, 2009]. The air sampled by ozonesondes, high alpine surface stations and regular aircraft (MOZAIC) are not identical, so some differences in measured O₃ concentrations are expected (e.g. high mountain sites can be affected by local wind systems, which remain to be analyzed in more detail). Nevertheless, it appears that the baseline O₃ evolution over Europe until around 2000 is probably more reliably described by high mountain sites and MOZAIC data than by the European ozonesondes. The records from the surface sites, aircraft and sondes show nearly consistent variability since 2000 compared to earlier times, which might point to some data quality problems in the earlier sonde data (Jennifer Logan, Department of Earth and Planetary Sciences, Harvard University, Cambridge, Massachusetts, USA, personal communication, 2009).

In summary, various studies have found significant positive temporal trends in the O_3 concentrations at northern temperate latitudes. However, there remains significant uncertainty regarding the magnitude and exact cause of these increases.

2.2.4. Implications for Surface Ozone Air Quality in Receptor Regions

Even though the O_3 surface concentrations on the downwind sides of the Atlantic and Pacific oceanic basins are the lowest found in the northern mid-latitudes, these concentrations still raise air quality concerns in regions receiving marine air inflow. These concerns arise both from human health and crop and ecosystem damage perspectives.

The concentrations of O_3 in air masses arriving at the Atlantic coastal fringes of Europe have been steadily rising over the past several decades (see Section 2.2.3), and have now reached levels

that exceed internationally accepted air quality guidelines and targets set for the protection of human health [50 ppb, maximum 8-hour level, *WHO*, 2006] and vegetation [40 ppb, accumulated time above 40 ppb, *Fuhrer et al.*, 1997]. Figure 2.9 presents the average monthly level of O_3 observed in baseline air masses arriving at Mace Head, Ireland taken from *Simmonds et al.* [2004] and extended up to July 2008. Monthly averages now exceed 40 ppb during much of the growing season in all years and approach or exceed 50 ppb in some spring months.



Figure 2.9. Monthly average O_3 mixing ratios in baseline air masses arriving at Mace Head, Ireland from 1987 – 2009, showing the 40 ppb and 50 ppb thresholds set for the protection of vegetation (orange line) and human health (red line), respectively.

As these baseline air masses travel further into the continent of Europe from the coastal fringes, surface uptake and photochemical destruction will necessarily erode the levels in Figure 2.9, whilst descent of ozone-rich air from above and photochemical production may supplement them. Figure 1.6 shows a model calculation of how a baseline air mass would further evolve during transport to southern England as a function of season; the sum of all contributions except Europe-regional represents the baseline O3 contribution at this further downwind site. Under these conditions, intercontinental transport will still represent a substantial fraction of any subsequent exceedance of air quality standards set in Europe for the protection of human health and vegetation.

The O₃ concentrations in air flowing into Japan have significant air quality impacts. Tanimoto [2009] reported the increase in the number of air quality standard exceedances for surface O₃ in spring at Mt. Happo (HPO), a rural mountain site in Japan ($36.7^{\circ}N$, $137.8^{\circ}E$, 1.85 km asl) that is largely influenced by long-range transport from the Eurasian continent. From 1999 to 2006 the number of days that O₃ exceeded 84 ppbv increased from 10 to 15 to about 30, approximately doubling over the course of 8 years. In 2006 HPO experienced O₃ levels exceeding 120 ppbv for the first time. This increase in the frequency of high-ozone episodes, which exceed the air quality standard thresholds, is likely due to significantly increased O₃ transport from the upwind regions. Trends in the tropospheric NO₂ columns over east-central China in spring indicate that region possibly contributes to the large O₃ increase at HPO.

The impact of the long-range transport of O_3 is also substantial in western North America on the eastern side of the Pacific Ocean. Air with low O_3 concentrations flows ashore within the marine boundary layer, but a strong vertical gradient is present above the boundary layer at the North American west coast. Ozonesondes launched from Trinidad Head, California, a coastal NOAA observatory (http://www.esrl.noaa.gov/gmd/obop/thd/), find that summertime O_3 mixing ratios average only 23 ± 7 ppbv (\pm one standard deviation) at 100 m, but are more than twice as high (53 ± 16 ppbv) at 2 km [*Oltmans et al.*, 2008]. As the onshore marine airflow encounters the complex topography that characterizes much of the North American west coast, ozone-rich air from above the marine boundary layer may be transported to the surface.

Some experimental analysis does indicate that transported baseline O_3 makes a major contribution to surface O_3 at some western North American sites, even during periods when the U.S. National Ambient Air Quality Standard (NAAQS) is exceeded. Jaffe et al. [2004] discuss an O_3 exceedance that occurred in the Seattle, Washington area due to long-range transport of fire emissions from Siberia. In addition, summertime surface O_3 in California's Central Valley directly inland from the Trinidad Head ozone sonde launch site often exceeds the NAAQS as indicated in Figure 2.10 [*Parrish et al.*, 2010]. The mean maximum 8-hr average O_3 on exceedance days (i.e. those days when 8-hr average O_3 exceeds 75 ppbv) is 20 ppbv higher than on non-exceedance days. The transported baseline O_3 , as measured 22 hours earlier by the Trinidad Head sondes, is also higher (by 11 ppbv) on exceedance days, which indicates that about half of the average excess O_3 on exceedance days is due to a larger baseline contribution. The remaining difference is presumably due to greater local and regional photochemical O_3 production. The average transported baseline O_3 on the exceedance days (59 ppbv) accounts for a very substantial fraction of the 75 ppbv standard.



Figure 2.10. Correlation between the interpolated daily maximum 8-hr surface O_3 averages at Tuscan Butte (a California surface site 200 km inland from the Pacific coast) with the O_3 mixing ratio measured by sondes 22 hours earlier at Trinidad Head. The sonde mixing ratios are averaged over altitudes from 1.0 to 2.5 km. The solid line shows the bi-variate linear, least-squares regression to the data, and the dashed line shows the standard regression.

2.2.5. Summary, Remaining Uncertainties and Future Needs

FINDING: Satellites clearly observe long-range transport of CO. Source gases with shorter lifetimes such as NO₂, are transformed more quickly in the atmosphere, making detection of transport difficult far from emission regions. Distinct signatures of long-range transport of O₃, which has both natural and anthropogenic sources, are difficult to detect by satellite, although significant O₃ enhancements are apparent over and downwind of regions with large precursor emissions. Present satellite retrievals are limited to tropospheric columns with little vertical profile information.

RECOMMENDATION: Further work is needed on quantitative satellite-based estimates of long-range transport of O₃, its inter-annual variability and its relation to changes in baseline O₃ concentrations.

FINDING: Measurements suggest that during the latter half of the 20th century, concentrations of O_3 at northern mid-latitudes increased by a factor of 2 or more. It is likely that much of this increase is due to increases in anthropogenic emissions of O_3 precursors. Within the limits of the measurement records, the increase has been comparable throughout all longitudes, and has occurred in all seasons. More recently, faster increases appear to have taken place downwind of eastern Asia, and the increase within the boundary layer of central Europe and North America has slowed.

RECOMMENDATION: The measurements documenting changes in surface and free tropospheric O_3 over the last several decades need to be systematically and collectively reviewed using consistent methods of analysis. Further, appropriate, systematic comparison with global models, which currently have problems accurately reproducing observed trends [e.g., *Lamarque et al.*, 2010], is required as an on-going activity.

FINDING: Measurements show that onshore marine airflow above the marine boundary layer can carry O_3 concentrations that approach urban air quality standards, and that air can mix to the surface and contribute substantially to air quality standard violations. This is particularly noticeable in low emission regions. The impact depends upon vertical mixing of air into the PBL, which is enhanced by complex topography.

RECOMMENDATION: Conduct focused research efforts that couple measurements with models to better quantify the process of air exchange between boundary layer and free troposphere in order to fully understand how free tropospheric O₃ enhanced by long-range pollution transport is mixed to surface.

2.3. Long-range Transport of Particulate Matter and its Precursors

Observations of haze layers in the Arctic during spring [*Shaw*, 1975], later attributed to transport from Eurasia [*Shaw*, 1995], and acid deposition in Canadian and Scandinavian lakes [*Likens and Bormann*, 1974] were the first recorded indications of the large-scale spatial transport of aerosol pollution. In the mid-1990s it became clear that long-range transport of PM could affect a downwind continent and significantly increase baseline concentrations [*Keating et al.*, 2005]. Since that time, a number of studies focused on regional PM levels strongly suggest that trans-boundary contributions dominate the observed concentrations in many areas. Development of new observing tools, improvements to the existing emission models and inventories, and a better understanding of chemical transformations and removal processes now provide better constraints on fundamental processes that drive the global distributions of the key aerosol species. Comprehensive studies that combine analysis of satellite-derived and ground-based observations with modelled back trajectories offer evidence that trends in emissions on one continent can affect PM trends thousands of kilometres downwind [*Karnieli et al.*, 2009].

Satellite images depict the transport of aerosols over thousands of kilometres for specific events and in time-averaged statistics. Transport mainly occurs in the free troposphere along the major pathways of long-range transport of gas pollutants. Direct evidence for transport at high altitude can be derived from lidar networks or high altitude ground-based stations [*Matthias et al.*, 2004]. It is believed that direct aerosol injection into elevated layers is not very efficient from anthropogenic sources in urban/industrial settings in contrast to forest fire plumes, which are convectively lofted above the ABL by the thermal energy of the fires [*Park et al.*, 2004].

2.3.1. Quantitative Estimates of Total Particulate Matter Transport from Satellites

Satellites provide a unique view of the transport of aerosols across ocean basins capturing specific events and aiding interpretation of in situ measurements. Time-averaged images of satellite-retrieved aerosol optical depth (AOD), a measure of total column PM loading, reveal the major pathways of aerosol transport from Asia across the Pacific Ocean to North America, and from Africa west across the Atlantic Ocean. At first such satellite imagery was only two dimensional, but recent analysis of CALIPSO (Cloud-Aerosol Lidar and Infrared Pathfinder Satellite Observations) observations is allowing depictions of three-dimensional aerosol transport [*Ben-Ami et al.*, 2009], e.g. Figure 2.11 [*Liu et al.*, 2008]. Altitude information is especially important for calculations of mass

flux and to determine what fraction of the PM seen from satellite are in the ABL, thereby affecting local air quality. Within the past few years, satellites have begun to provide quantitative information on intercontinental aerosol transport. The Multiangle Imaging SpectroRadiometer (MISR) is able to characterize AOD, size and degree of nonsphericity. Figure 2.12 shows results from a study [*Kalashnikova and Kahn*, 2008] where particle loading, size and nonsphericity all decreased during transport across the Atlantic.



Figure 2.11. A dust event that originated in the Sahara desert on 17 August 2007 and was transported to the Gulf of Mexico. Red lines represent back trajectories indicating the transport track of the dust event. Vertical images are 532 nm attenuated backscatter coefficients measured by CALIPSO when passing over the dust transport track. The letter __D''designates the dust layer, and _S'' represents smoke layers from biomass burning in Africa (17–19 August) and South America (22 August). Graphic courtesy of Kurt Severance at NASA Langley Research Center.

Following Kaufman et al. [2005], who applied a quantitative method to determine the net transport of dust from Africa to the Amazon basin, Yu et al. [2008] have used MODerate resolution Imaging Spectroradiometer (MODIS) aerosol products to determine the transport of —pllution" PM across the Pacific Basin. They isolated the pollution portion of the aerosol optical thickness measured by MODIS and translated the optical thickness to total column mass using a relationship derived from field experiments. Analyzed wind fields then allowed a quantitative calculation of the amount of pollution PM mass leaving the Asian continent and the amount arriving at North America (Figure 2.13). MODIS observed about 18 Tg/yr leaving Asia within the latitude range of 30 to 60°N and 4.4 Tg/yr arriving in North America. This compares well with results from the Goddard Global Ozone Chemistry Aerosol Radiation Transport (GOCART) and Global Modelling Initiative (GMI) chemical transport models where the model PM was defined as the sum of the BC, organic material and sulphate. The MODIS data show a seasonal cycle with a transport maximum in spring, but transport is observed in all seasons. Uncertainty analysis showed that the estimation of the aerosol transport altitude contributes the largest uncertainty to the satellite-based estimate.



Figure 2.12. Evolution of dust plume characteristics for four case studies of dust transported from Africa to the Caribbean across the Atlantic in six days. Top: AOD, a total column measure of PM loading, decreases due to dry deposition. Center: Angstrom Exponent, an inverse measure of particle size. Particles on Day 6 are smaller than on Day 1 due to gravitational settling of the larger particles. Bottom: Particle sphericity showing less nonspherical fraction at the end of the transport due to increase in the hygroscopicity of particles and/or the removal of dust. [Reprinted from Figure 11 in Kalashnikova, O. V., and R. A. Kahn (2008), Mineral dust plume evolution over the Atlantic from MISR and MODIS aerosol retrievals, Journal of Geophysical Research, 113(D24204).]

Figure 2.13. Estimates of annual pollution aerosol flux leaving Asia in the West Pacific (a) and arriving in North America in the East Pacific (b) for different latitude ranges. The blue bars are derived from MODIS aerosol products, and the red and green bars are derived from model calculations. [Reprinted from Figure 7 in Yu, H., et al. (2008), A satellite-based assessment of transpacific transport of pollution aerosol, *Journal of Geophysical Research, 113*(D14S12).]

2.3.2. In Situ and Lidar Observation of Particulate Matter Outflow from Continents

The outflow of PM and its precursors from mainland Asia and North America has been studied from downwind island sites. Bermuda, a site of long-term measurements from the AEROCE (Atmospheric/Ocean Chemistry Experiment) campaign [*Prospero*, 2001], has provided much information on the outflow of O_3 , PM and their precursors from North America. In Asia, dust and

anthropogenic PM transported from the mainland have been observed in Japan. Figure 2.14 illustrates one event with two peaks evident due to anthropogenic PM. The elemental carbon/organic carbon (EC/OC) ratio remained at 0.2 during the whole episode, which indicates anthropogenic PM was present even when dust dominated [*Takami et al.*, 2006]. Episodes of enhanced SO₂ concentrations measured at the summit of Mt. Fuji, Japan (3.8 km asl) provide another example of Asian outflow. Enhanced CO and radon (²²²Rn) concentrations always accompanied the enhanced SO₂ concentrations, and backward trajectories for such events indicate an Asian continental origin [*Igarashi et al.*, 2006].



Figure 2.14. Aerosol transport event observed at Cape Hedo, a sea level site at the northern tip of Okinawa Japan. From top: (a) sulphate and organic components; (b) organic carbon, elemental carbon and PM2.5; and vertical distributions of (c) dust (non-spherical particles) and (d) deliquesced anthropogenic aerosol (spherical particles). [Reprinted from Takami, A., et al. (2006), Transport of anthropogenic and dust aerosol observed at Cape Hedo, Okinara, *Journal of Aerosol Research, 21*(4): 341-347. Copyright 2006 Japan Association of Aerosol Science and Technology.]

2.3.3. Observations of Particulate Matter in Continental Inflow

Measurements on the eastern sides of the North Atlantic and North Pacific Oceans record the arrival of pollution PM from North America and Asia, respectively. At Mace Head, Ireland, anthropogenic sulphate has accounted for 85-90% of the total non-sea salt sulphate (nss) sulphate during marine inflow conditions, indicating that the aerosol arrived from across the ocean [*Savoie et al.*, 2002]. The nss sulphate values at Mace Head are several times greater than found at similar latitudes in the Southern Hemisphere, providing further evidence for an anthropogenic source (Figure 2.15) [*Barrie et al.*, 2001]. The ICARTT campaigns provided information on pollution transport across the North Atlantic Ocean. Putaud et al. [2004] and Van Dingenen et al. [2004] provided an extended review of physical and chemical properties of PM in Europe. It is shown that the regional background in Europe is

 7.0 ± 4.1 micrograms per cubic meter ($\mu g/m^3$) for PM₁₀ and $4.8 \pm 2.4 \mu g/m^3$ for PM_{2.5} and that this background is not only due to natural sources but also to long-range transport of anthropogenic PM.

Because forest fire emissions can be injected into the upper troposphere, forest fire aerosols are efficiently transported over long-distances. Aerosol layers transported from North America to Europe have been captured by lidars over Western and Central Europe [*Forster et al.*, 2001; *Wandinger et al.*, 2004] and by surface measurement of BC concentrations. Analyses from EARLINET (European Aerosol Research Lidar Network) have shown that aerosol plumes from the anthropogenic sources in the North American boundary layer are generally not similarly intense [*EARLINET*, 2003]. Other examples of long-range transport of fire plumes include smoke originating from the Ural region travelling both northwest towards Scandinavia and eastward, across Canada, Scandinavia and eastern Europe before returning to Russia in a little more than 2 weeks [*Damoah et al.*, 2004].



Figure 2.15. Average sulphate levels measured at marine sites in the Northern and Southern hemispheres [*Barrie et al.*, 2001].

Measurements also demonstrate that Asian industrial sources account for at least some of the baseline PM concentrations that are observed at the west coast of North America [Wilkening et al., 2000]. Figure 2.16 illustrates an episode of elevated sub-micron aerosol scattering that corresponds to approximately 20 μ g/m³ of PM₁₀. The observed Hg⁰/CO ratio in this episode indicates that the detected aerosol was associated with Asian industrial sources, not mineral dust or biomass burning [Jaffe et al., 2005b; Weiss-Penzias et al., 2007]. Using surface aerosol measurements from the Interagency Monitoring for Protected Visual Environments (IMPROVE) measurement sites together with a global chemical transport model. Heald et al. [2006] estimate the mean Asian sulphate enhancement in the northwestern U.S. in spring is $0.16 \pm 0.08 \,\mu\text{g/m}^3$. This is much smaller than the annual average ambient air quality standard of 15 μ g/m³ for PM_{2.5}, but is higher than the estimated natural concentration of 0.09 $\mu g/m^3$ presently used as the objective for regulating visibility in U.S. wilderness areas. Interestingly, no such Asian enhancements are observed for nitrate or for organic carbon (OC) aerosol. A significant sulphate contribution from Asian emissions is supported by the recent study of van Donkelaar et al. [2008] as part of the INTEX-B campaign, which found that 56% of the measured sulphate between 500– 900 hectopascals over British Columbia is due to East Asian sources, and that there has been a 72–85% increase in the relative contribution of East Asian sulphate to the total burden in spring off the northwest coast of the U.S. since 1985. The ITCT-2K2 campaign observed additional evidence of trans-Pacific transport of anthropogenic Asian aerosols. Figure 2.17 illustrates PM measurements made during an aircraft profile through layered plumes marked by variations in CO concentrations. Transport modelling identified Asia as the emission source region. Another ITCT-2K2 flight observed particle size distributions and sulphate mass concentrations consistent with nearly pure sulphuric acid particle formation in the central North Pacific Ocean. The transport of gas-phase precursors from Asia and formation of particles in the mid-Pacific avoids the scavenging in cyclonic systems that remove PM from the atmosphere. Such mid-oceanic PM production suggests a larger potential for particle pollution to arrive during intercontinental transport events [Brock et al., 2004].

BC, defined as the light-absorbing fraction of carbonaceous aerosols, has complex climatic implications involving atmospheric heating and snow and ice surface warming. BC is difficult to sense remotely due to its small contribution to PM mass and aerosol optical depth. However its absorbing properties allow detection in the aerosol index product from Total Ozone Mapping Spectrometer (TOMS) [*Torres et al.*, 2002] or more recently OMI [*Torres et al.*, 2007]. Since these products are most sensitive to high-altitude absorbing aerosols, they are most useful for detecting biomass burning aerosols, but they are also somewhat sensitive to dust outflow. To some extent long-range transport of black carbon can also be detected using the absorption aerosol optical depth (AAOD) products from AERONET (AErosol RObotic NETwork) [*Dubovik et al.*, 2002], with the caveat that this product is sensitive to absorbing dust as well as black carbon. Atlantic and Pacific outflow regions have relatively large AAOD (0.005 to 0.01), and high latitude sites such as Bonanza Creek Alaska and Tomsk, Russia have substantial AAOD (0.01 to 0.02) similar to levels characteristic of Europe.



Figure 2.16. Concentrated trans-Pacific pollutant transport event observed at a mountaintop site near the west coast of North America. Observations of O_3 , aerosols, Hg and CO in a plume transported from Asia to the Mt. Bachelor Observatory in Oregon near the west coast of the U,S, in April 2004. The O_3 vs. CO linear correlation gives an R^2 value of 0.8 and a slope of 0.22. [Reprinted from Figure 4 in Jaffe, D. A., et al. (2005b), Export of atmospheric mercury from Asia, *Atmospheric Environment*, *39*(17): 3029-3038, with permission from Elsevier.]

Better understanding of BC long-range transport requires improved quantification of the vertical distribution of BC. With the development of the SP2 instrument, high quality BC measurements with high vertical resolution are becoming available from aircraft campaigns [e.g. *Schwarz et al.*, 2006]. Aircraft campaigns near Houston and Costa Rica [*Schwarz et al.*, 2006; *Schwarz et al.*, 2008] indicate that BC concentrations decrease with altitude by about two orders of magnitude between the boundary layer and the upper free troposphere, which implies efficient scavenging of BC during vertical transport in these regions. On the other hand, aircraft SP2 measurements indicate substantial BC pollution in the Arctic, with increasing concentrations within the lower troposphere [*Koch et al.*, 2009, Figures 9 and 10]. This view is broadly consistent with the AERONET and OMI perspective of relatively clean BC conditions in the remote lower latitudes of the northern hemisphere but substantial pollution at higher latitudes. The substantial pollution occurs in spite of decreasing BC trends in recent decades in North American surface measurements [*Sharma et al.*, 2006] and snow [*Hegg et al.*, 2009], but with snow concentrations in Eurasia 2-3 times those in North American snow. As future campaigns measure BC in the free troposphere, understanding of long-range BC transport should greatly improve.



Figure 2.17. Aerosol measurements marked by variation in CO concentrations. Vertical distributions of CO with aerosol particle number size distribution (left) and particle volume size distribution (right) in a plume of emissions transported from Asia. The measurements were made near the west coast of North America on 5 May 2002. [Adapted from Figure 2 in Brock, C. A., et al. (2004), Particle characteristics following cloud-modified transport from Asia to North America, *Journal of Geophysical Research, 109*(D23S26), doi:10.1029/2003JD004198.]

2.3.4. Trends in Surface site Observations of Particulate Matter

As discussed in previous sections, satellite remote sensing observations can clearly identify the intercontinental transport of PM. Complementary in-situ surface measurements have revealed that the long-range transport events could have significant impact on surface PM loading in receptor regions. Furthermore, the analysis of the long-term trend observed in the receptor regions provides an effective measure of the —integated effect" of the transport processes in larger spatial and temporal scales, e.g., the long-range transport influence on the receptor regional-scale PM loading and baseline concentrations. In this section, we discuss the long-term trends observed in receptor regions, by both in-situ and remote sensing measurements, under the influence of the transport from the East Asian, North American, and European sources. In addition, comparisons are made between the observed long-term trend and the emission trends in the source regions.

Analyses of measurements of nitrate and anthropogenic nss sulphate concentrations on island sites in the Pacific and the Atlantic provide temporal trends, which show the effect of the long-range transport of these pollutants from the East Asian, European, and North American source regions. The trend in nss sulphate pollution is estimated by computing the "natural" nss-SO₄⁼ based on the methanesulfonic acid concentrations and subtracting from the total nss sulphate. On Midway Island anthropogenic sulphate concentrations approximately doubled from 1981 to the mid-1990s [*Prospero et al.*, 2003], a trend that closely matches the increase in SO₂ emissions from China over that period [*Streets et al.*, 2000]. Nitrate concentrations yield a similar trend over the same time period. The measurements made in Bermuda show coherent patterns in the annual mean concentrations of nss sulphate and nitrate (Figure 2.18). The nss sulphate decreased steadily from the start of the measurements until the mid-1990s, then stabilized and finally increased slightly through the remainder of the period. The emissions of SO₂ from U.S. sources decreased substantially over this time period by an amount that roughly corresponds to the nss sulphate trend observed at Bermuda, yet the timing of the decreases are markedly different; the U.S. SO_2 emissions decreased sharply between 1994 and 1995 when more stringent controls were mandated. In contrast, there is no substantial change in nitrate over this period, in agreement with the trend in NO_x emissions, which shows only a very slight increase over the period. The substantial differences between the aerosol record and the emissions record demonstrate the challenges in attempting to monitor long-term trends at distant receptor sites.

Aerosol sulphate and nitrate measurements have also been made continuously on Barbados since 1989. This record suggests that nitrate remained unchanged and that nss-SO₄⁼ decreased by about 20%. Approximately half of the nitrate and nss sulphate at Barbados is attributable to anthropogenic sources [*Savoie et al.*, 2002], predominately from European sources [*Hamelin et al.*, 1989]. If one assumes that the change in nss sulphate concentration is due to the transport of pollutants and if one considers that about half of the nss sulphate is natural (from DMS), then the actual decrease in sulphate pollution is roughly 40%, a change that is consistent with the sharp drop in European SO₂ emissions over this time period [*EMEP*, 2004]. However the absence of a discernable change in Barbados nitrate concentrations is puzzling in light of the substantial reduction of European emissions of NO_x, roughly 25% [*EMEP*, 2004].



Figure 2.18. Annual mean nss-sulphate and nitrate concentrations on Bermuda during onshore winds compared to eastern US SO₂ (Tg S/y) and NOx emissions (Tg N/yr). Bars indicate the standard deviations of the monthly means from which the annual means are derived. [Based on data discussed in part in *Savoie et al.*, 2002]

The phenomenon of Arctic Haze occurs each winter and early spring due to rapid, meridional transport of anthropogenic PM from the northern mid-latitudes to the Arctic. The result is a dramatic increase in aerosol mass concentration. The haze is composed of a varying mixture of sulphate, organics, ammonium, nitrate, dust, and BC [e.g., *Li and Barrie*, 1993; *Quinn et al.*, 2002]. It is also rich in distinct heavy metals, allowing for the identification of particular industrial sources [e.g., *Rahn and McCaffrey*, 1980]. Long term trends in the aerosol chemical composition and optical properties of Arctic Haze have been monitored at stations within the Arctic including Barrow, Alaska (71.3°N, 156.8°W); Alert, Canada (82.5°N, 62.3W); and Station Nord, Greenland (81.4°N, 16.6°W).

The longest record of sulphate concentrations in the Arctic (1980 to present at Alert) reveals no change in sulphate concentration during the 1980s [*Sirois and Barrie*, 1999] followed by a decline beginning in 1991. A similar decline through the 1990s was observed in sulphate concentrations at Station Nord [*Heidam et al.*, 2004] and in light scattering and absorption at Barrow [*Bodhaine and Dutton*, 1993]. A combined modelling and measurement analysis of sulphate concentrations at Station Nord was able to account for scatter in measured concentrations due to changing meteorology [*Heidam et al.*, 2004]. With the meteorological variability removed, it was possible to attribute the decrease in concentration to a reduction in emissions. The model that was used estimates that more than 70% of the sulphur measured at Station Nord is emitted from the region making up the former Soviet Union. This result is supported by a 50% decrease in Russian sulphur emissions reported to EMEP during the 1990s [*Vestreng*, 2003]. It is not clear from this analysis how reductions from Europe and North America influenced the sulphate concentrations at Station Nord.

The decreasing trend in sulphate at Alert detected through the 1990s has continued into the present century [*Quinn et al.*, 2007]. In contrast to sulphate, nitrate concentrations have increased during the Arctic Haze season at Alert over the period from 1981 to 2003. This increase may be a result of increasing vehicle usage in regions of the former Soviet Union. Quinn et al. [2009] compared ratios of tracer species measured at Barrow during the Arctic Haze season from 1976-1977 and 1997-2008 to assess trends in Arctic Haze aerosol over this 30-year period and changes in source regions. The tracer species considered in the analysis included nss sulphate (derived from fossil fuel combustion), non-crustal (nc) vanadium (V) (derived from the combustion of heavy residual oil), and nc manganese (Mn) (derived from iron, steel, and ferro-alloy manufacturing, coal and oil combustion, and mining). Based on these two data sets, concentrations of nss sulphate and nc V have decreased at Barrow by about 60% over this 30 year period. Consistency in the ratios of nss sulphate to nc V and nc Mn to nc V between the two data sets indicates that, while emissions have decreased in source regions, the source regions themselves have remained the same over this time period.

Satellites have been used to estimate temporal changes of aerosols through measurements of the AOD, which is a good indication of aerosol loading. Satellite AOD measurements provide the most comprehensive view of the atmospheric aerosols on global and regional scales. The available satellite AOD data products span nearly 2 decades. Mishchenko et al. [2007] analyzed the temporal variation in the globally averaged GACP (Global Aerosol Climatology Project) AOD data, which were derived from Advanced Very High Resolution Radiometer (AVHRR) radiance measurements. The GACP AOD showed a small increase, i.e., 0.03 or ~3% over 14 years, but a statistically significant decrease from 1991 to 2005. More recently, Thomas et al. [2010] found an increase in global AOD of 0.024/decade between 1995 and 2001. This study was based on the Global Retrieval of ATSR Cloud Parameters and Evaluation (GRAPE) dataset, which was derived from ATSR-2 (Along Track Scanning Radiometer-2) observations. These authors attributed the global GRAPE increase to regional increases over the North Pacific, Tropical Pacific, and Tropical Indian oceans. It was also noted that there were significant differences between the GACP and GRAPE AOD data, approaching 0.5, and both datasets showed significantly larger shorter-term variations (uncorrelated) than the derived long-term trend. Li et al. [2009] presented a more extensive comparison of the satellite AOD data, including MISR, MODIS, and GACP (Figure 2.19). This comparison also showed a considerable difference between the AOD datasets in terms of absolute magnitude as well as shortterm and long-term temporal variations. Again, the shorter-term variations are much larger that the long-term changes. These authors concluded that most of the current satellite observation-based AOD are inadequate for analysis of long-term changes without significant improvement in calibration/validation, cloud screening, characterization of aerosol microphysical and optical properties, and surface reflectance.

2.3.5. Implications for Surface Particulate Matter Air Quality in Receptor Regions

Particles transported over long distances can have a negative impact on urban air quality, although observed events are generally limited to dust transport. Figure 2.20 illustrates a particularly pronounced event. The dust was transported initially in the free troposphere, and then brought to the surface through a combination of processes, which differed from region to region. Subsequent mixing with local pollutants led to $PM_{2.5}$ concentrations exceeding the new U.S. daily standard of 35 µg/m³. During this specific case, the dust passed above the Pacific Northwest (i.e. Seattle, Washington) and had greater impact at the surface in the Rocky Mountains and eastern United States. Even in events when the percent increase in concentration is small, it may be sufficient to cause local exceedances of air quality standards.



Figure 2.19. Comparison of global mean AOD over ocean derived from different satellite input data and retrieval algorithms for an overlapping period since 2000. Different retrieval algorithms assume different aerosol models. [Reprinted from Figure 1 in Li, Z., et al. (2009), Uncertainties in satellite remote sensing of aerosols and impact on monitoring its long-term trend: A review and perspective, *Annales Geophysicae*, *27*: 2755-2770.]



Figure 2.20. Contributions from three sources to surface $PM_{2.5}$ in three U.S. cities (Salt Lake City, Utah; Atlanta, Georgia and Seattle, Washington) during an April 2001 episode of Asian dust transported to North America. [Adapted from Jaffe, D., et al. [2003], The 2001 Asian Dust Events: Transport and Impact on Surface Aerosol Concentrations in the U.S., EOS: *Transactions of the American Geophysical Union, 84*(46): 501-507.]

Until recently, satellite observational tools were unable to provide robust source apportionments. The added capability of CALIPSO may significantly improve quantitative estimates of PM flux and the impact of transported particles on air quality at ground level over broad regions. However, even the enhanced observational capabilities are insufficient to provide robust quantitative air quality assessments alone. To provide an answer, improved observational capabilities will need to be combined with modelling in a synergistic approach.

2.3.6 Summary, Remaining Uncertainties and Future Needs

FINDING: In situ measurements have established the importance of intercontinental transport of PM from dust, forest fires, and anthropogenic sources. In some case studies of in situ measurements have illuminated secondary aerosol formation far from the sources of transported precursors. FINDING: Ground-based lidar networks and mountain top measurement sites in Europe, North America and Asia provide large continuous data sets that characterize the frequency and intensity of aerosol transport events and the meteorological conditions responsible for them. They also provide important information on aerosol particle properties. Operational procedures for dust outbreak monitoring in near real time are implemented at several mountain sites in Europe.

RECOMMENDATION: Discussion is required regarding the optimum observation strategy for in situ and surface remote measurements for further characterizing intercontinental transport of aerosols. A particular focus should be on measurements to quantify the sources and properties of the organic and black carbon components of transported PM, which are currently poorly understood. The mechanisms for the transport and removal of PM also need to be addressed.

FINDING: Over the last few years, satellites have begun to provide quantitative information on intercontinental aerosol transport, including estimates of the amount of dust and pollution transported, the altitude of transport and, in some cases, aerosol particle properties.

FINDING: Observed trends of PM (e.g. SO₄) in downwind locations are closely related to the emission trends in the source regions with the impact of long-range transport more evident in remote regions less affected by local sources such as the Arctic. Satellite measurements are currently not precise and accurate enough for monitoring the long-term changes.

RECOMMENDATION: Analysis of available datasets should be supported to better quantify aerosol particle properties and the PM fluxes (pollution, dust, biomass-burning emissions) between continents in the Northern Hemisphere and to/from regions to the north (Arctic) and south (e.g. Africa). Surface in situ and remote measurements as well as satellite observations can provide important contributions.

FINDING: Long-range transport of aerosols can have air quality and climatic implications, at northern latitudes especially in the outflow from the Asian continent and in the Arctic region. Observational studies alone have not well quantified either the impact on local air quality or the climatic implications.

RECOMMENDATION: To inform air quality control decisions, better methodologies are required to extract the long-range transport contribution from measured overall PM concentrations. Characterizing long-range transport of black carbon is particularly important for understanding climatic implications.

2.4. Observational Evidence for Attribution of Source Regions

Enhancements in trace gas and aerosol concentrations associated with long-range transport of pollution from sources upstream may persist for several days in the free troposphere, where residence times are enhanced. During transport, atmospheric mixing acts to dilute these enhancements, as air masses of different histories are mixed together creating a spectrum of chemical signatures downstream from the source region. This provides a challenge in attributing source regions of longrange transport events using observations from research aircraft and surface stations, since discrete chemical signatures associated with source regions may be completely masked by mixing. Nevertheless, observed relationships among chemical tracers can be used to detect the influence of upstream source regions on the composition of a given region of the atmosphere. Knowledge of source relationships between tracers and their atmospheric processing, and observations of tracers of different atmospheric lifetimes, can be used to yield estimates of the influence of remote upstream sources on atmospheric composition. Moreover, methods for combining in situ or satellite tracer observations with backward modelling techniques provide powerful constraints on the pathways of long-range pollution transport, our understanding of chemical processing during individual transport events and attribution of contributions from different source regions and types. Improvements in the quality of analyzed wind fields from meteorological services has also enabled more robust attribution of contributing source regions to a given receptor region downstream.

2.4.1. Meteorologically-based source attribution studies

On the intercontinental or hemispheric scale, meteorology-based analyses for accurately determining the emission source regions of gases or PM at a particular receptor require the use of transport models. These modelling techniques currently fall into two categories: 1) backward simulations with Lagrangian particle dispersion models (LPDM); and 2) adjoints to chemical transport models (CTM). These methods are designed for exploring emission regions (both known and unknown) and transport pathways backwards in time from an in situ trace gas or PM measurements. They are also complementary to the more traditional approach of using a model to emit tracers from known emission sources and seeing if they advect forwards in time to the receptor location.

LPDM backward simulations initialized from a receptor (measurement) location is the now the most commonly used method of meteorological-based source attribution studies. Simpler back trajectories are still used but they are limited in the quantitative information they can provide on the contribution from a source region, and do not perform as well in regions of strong shear or convection. LPDMs calculate the transport pathways of air masses (often referred to as <u>particles</u>" because they are assumed to be infinitesimally small) following the winds resolved in atmospheric analyses or forecasts. The models also include a stochastic step to represent the effects of unresolved turbulence and convection [*Stohl et al.*, 2002]. The parameterised <u>random walk</u>" shuffles particles, each weighted with the same tracer mass, so that their sum within a volume represents the effects of advection and diffusive mixing on passive tracer concentration [*Legras et al.*, 2003]. At present, LPDMs have not yet been developed to include chemical or aerosol processing or detailed descriptions of deposition processes where non-linearities in chemical transformation may be important.

For tracers, such as CO where a linear loss can be included, LPDM simulations lead to a relatively accurate representation of transport which takes into account the shear, turbulence and convection encountered by a volume of air as it travels from source to receptor. The dynamics of the atmosphere also conspire to allow accurate simulation of tracer structures formed by advection on scales an order of magnitude finer than the resolution of the wind field [Methven and Hoskins, 1999] allowing simulations at high resolution (< 0.5 degrees). LPDM backward simulations have become known as retroplumes [Stohl et al., 2003] which can be calculated in various forms and resolutions by models such as FLEXPART [Stohl et al., 2005], STILT [Gerbig et al., 2003; Lin et al., 2003] or HYSPLIT (http://www.arl.noaa.gov/HYSPLIT info.php). The method involves releasing a multitude (often tens of thousands) of trajectory particles in the receptor volume and tracking their location for several days backwards in time using high resolution (1° x 1° or finer) global wind fields. The sum of the residence time of all particles in the boundary layer establishes the —fotprint" of the retroplume and indicates the surface emission region influencing the measured trace gas or PM. Multiplying the footprint residence time by co-located surface emission rates estimates the contribution of that source region to the trace gas quantity measured at the receptor. One of the first examples of the use of the retroplume technique for source attribution of a long-range transport event, demonstrated that aircraftbased measurements of enhanced CO and NO_v in the free troposphere above Europe during November, 2001, originated in the boundary layer of North America and travelled to Europe via a warm conveyor belt [Stohl et al., 2003]. Approximately half of the CO in the plume came from various anthropogenic sources in the eastern U.S., with the main contribution coming from the New York region. The technique has also been used to show how elevated O_3 and CO above the California coast originated in a broad region of south and east Asia with subsequent transport to California via two successive warm conveyor belts [Cooper et al., 2004]. Retroplumes have also shown how Asian pollution can be rapidly transported eastwards in the upper troposphere to Europe in as little as 6 days [Stohl et al., 2007b]. Application to aircraft and surface measurements in the Arctic has recently revealed the pathways of biomass burning emissions from Europe [Stohl et al., 2007a] and central Asia [Warneke et al., 2007] to Spitzbergen and northern Alaska, respectively. A recent development in this technique is to calculate retroplume climatologies for thousands of in situ measurements at a given location over many years. The purpose is to identify the typical transport patterns and source regions associated with a subset of measurements such as enhanced or depleted trace gas or PM events [Cooper, 2010; Hirdman et al., 2010].

Meteorologically-based source attribution studies can also be conducted with CTM adjoints. While adjoint capabilities have been around slightly longer than the retroplume technique, they have not been as widely applied to intercontinental- or hemispheric-scale studies, although they are gaining in popularity. The adjoint works in the same spatial and temporal resolution as the CTM to which it is attached and provides similar output to retroplumes, identifying the source region of an air mass at a particular receptor as well as its transport pathway. On the hemispheric scale, adjoint studies have a coarser horizontal resolution than the retroplume studies (roughly $2^{\circ} \times 2.5^{\circ}$ vs. 1° x 1°). The advantage of adjoints over retroplumes is that they can be used to not only determine the source region of a trace gas or PM measurement, but can also identify the chemical and physical mechanisms that modify the trace gas or PM during transport, provided these mechanisms are handled by the CTM.

Application of adjoints to pollution plume studies began with the development of the adjoint to the HANK CTM which demonstrated that the techniques could identify plume transport from Asia to Mauna Loa Observatory, Hawaii [*Vukicevic and Hess*, 2000]. Adjoints have been recently applied to explore processes that influence intercontinental transport to North America. Henze et al. [2009] used the GEOS-Chem adjoint to determine the source regions of inorganic PM_{2.5} precursor emissions for monitoring sites throughout the U.S. and found that intercontinental influences are small, though transboundary influences within North America are significant, with strong influence from SO_x emissions in Mexico. Zhang et al. [2009] determined that Mount Bachelor Observatory in the northwestern U.S. experiences distinct Asian O₃ pollution episodes, with most of the O₃ production occurring over East Asia, adding to a diffuse background production distributed over the extratropical northern hemisphere (Figure 2.21). The adjoint also showed that transpacific pollution plumes transported in the free troposphere are diluted by a factor of 3 when entrained into the boundary layer, explaining why these plumes are undetectable in U.S. surface air.



Figure 2.21. Measured O_3 mixing ratios of 55-60 ppbv at Mt. Bachelor Observatory, Oregon on 1 May, 2006, were determined as having a 50% contribution from Asian sources based on the global GEOS-CHEM model. The model adjoint showed that much of the O_3 production over the past 60 days occurred within Asia (left panel shows where O_3 production occurred) roughly 10 days earlier (right panel shows when O_3 production occurred in 4 different regions over the previous 60 days), although production also occurred as the Asian pollution plume crossed the North Pacific Ocean and descended into the mid-troposphere over North America. [Reprinted from Figure 2a in Zhang, L., et al. (2009), Intercontinental source attribution of ozone pollution at western U.S. sites using an adjoint method, *Geophysical Research Letters*, *36*(L11810).]

FINDING: High resolution particle dispersion models provide powerful techniques for the identification of pollutant source regions.

FINDING: Recent developments in adjoint modelling techniques provide useful insights into the sources and the chemical transformation processes that influence trace gases and aerosols during long-range transport.

RECOMMENDATION: There is a need for further application of particle dispersion models to the analysis of long-time series of ozone and PM, with the goal of determining the impact of source regions on long-term trends and inter-annual variability.

RECOMMENDATION: Development of LPDMs to include full treatments of chemical and aerosol processing and wet/dry deposition as a complement to Eulerian models is needed.

2.4.2. Source attribution based on trace gas correlations and ratios

Relationships among atmospheric chemical tracers are powerful observation-based tools for assessing source contributions and probing chemical and physical processing in air masses undergoing long-range transport. For example, the ratio ΔO_3 : ΔCO_3 , used as an estimate of the excess in O₃ and CO above background in different plumes, has been used to identify long-range transport events and estimate photochemical O₃ enhancement in polluted air masses downstream of the continents [Bertschi et al., 2004; Parrish et al., 1993]. ANOY: ACO has been used as a measure of export efficiencies of anthropogenic emissions from continental regions [Li et al., 2004; Stohl et al., 2002; Val Martin et al., 2006] and correlations between O₃ and PAN in aged air can be used to infer photochemical influence from sources upstream [Parrish et al., 1992]. These trace gas relationships can also be useful in distinguishing plumes from different sources (e.g. boreal forest fires versus anthropogenic pollution), however, care has to be taken when using ΔO_3 : ΔCO_3 to infer O_3 production. Secondary production of CO from non-methane volatile organic compound oxidation and O₃ deposition or photochemical O₃ loss during transport may modify observed slopes [*Chin et al.*, 1994], and photochemical destruction of CO may increase the ΔO_3 : ΔCO slope [*Real et al.*, 2010]. Tracer variability within an air mass and sharp tracer gradients at its edges may also complicate the use of ratios, where observations are non-representative of the plume average or are affected by background air at the point of sampling. Since plumes do not remain isolated, in reality their chemical signatures are influenced by the history of different backgrounds and source signatures with which they have mixed during transport. Price et al. [2004] showed ΔO_3 : ΔCO slopes in plumes transported in the upper troposphere were likely influenced by mixing with higher ozone concentrations. In addition, the choice of criteria used to define ΔO_3 : ΔCO above background can result in differences in slope values [*Pfister et al.*, 2006] and inference of plume source and photochemical activity. On a larger scale, correlations between satellite observations of tropospheric O_3 and CO over the Pacific have been used to identify and track events of trans-Pacific photochemical O₃ transport [Zhang et al., 2008].

The increasingly routine observation of a range of non-methane hydrocarbons (NMHC) during airborne missions and at free tropospheric surface sites is proving valuable for identifying and examining long-range transport events in the troposphere. Many simple alkane and alkene species have atmospheric lifetimes useful for probing processing on timescales for intercontinental transport (2-5 days). The robust source relationships among different NMHCs, their range of typical atmospheric lifetimes, and their common chief sink (the OH radical) have been used to infer photochemical ageing and OH concentrations from NMHC ratio observations during long-range transport events [Parrish et al., 1992; Price et al., 2004]. Atmospheric mixing destroys the idealised evolution of tracer ratios expected through only photochemical processing of an air mass, which can inhibit their use in source attribution or inference of plume processing. Nevertheless, results from the ICARTT pseudo-Lagrangian experiment (see 2.4.4) showed that on spatial and temporal scales of intercontinental transport, well-defined NMHC _fingerprints' can be retained in air masses over several days in the free troposphere. These NMHC fingerprints allowed successful pseudo-Lagrangian links to be identified. In addition, they provided a powerful independent validation of atmospheric transport model derived links among observation platforms. Lewis et al. [2007] demonstrated distinct NMHC signatures of large enhancements in acetylene relative to acetaldehyde and acetone in biomass burning plumes intercepted near the Azores compared with North American anthropogenic plumes, enabling robust separation of these two source types. The use of NMHC ratios in conjunction with ΔO_3 estimates can also be used to investigate relationships between O_3 photochemical production and plume age [Helmig et al., 2008]. Using statistical inference techniques applied to observed NMHC ratios in the ICARTT pseudo-Lagrangian air masses, Arnold et al. [2007] estimated mean OH concentration, dilution rates and background air mass NMHC composition, characteristic of air masses of anthropogenic and biomass burning origin in the summertime North Atlantic troposphere using robust relationships among NMHCs with a wide range of lifetimes, such that their OH oxidation during the 1-3 days between export from North America and aircraft interception produced an

appreciable change in NMHC ratios. Combination of NMHC ratios with Lagrangian particle dispersion model simulations has also allowed quantification of the different contribution of photochemistry, emissions and mixing [*Honrath et al.*, 2008; *Parrish et al.*, 2007].

Nevertheless, recent studies have indicated that other oxidants such as halogens (chlorine, bromine radicals) or nitrate may play an important role in the formation (or destruction) of tropospheric O₃. For example, chemical measurements in coastal regions of the Gulf of Mexico during the summer 2006 TexAQS study for the first time confirmed that nitryl chloride is a key atmospheric species that couples the nitrogen and halogen chemistries of the marine boundary layer [*Osthoff et al.*, 2008]. The use of inert tracers (e.g. perfluorocarbons), which have no chemical sinks, has also been used recently as part of the DLR POLARCAT-GRACE campaign to follow air masses over a few days providing important constraints on atmospheric mixing among different air masses (Hans Schlager, Deutsches Zentrum fuer Luft - und Raumfahrt (DLR) Institut fuer Physik der Atmosphaere, Wessling, Germany, personal communication, 2010).

FINDING: Increasingly routine observations of concentrations of ozone and aerosol precursors as well as reservoir species such as PAN, onboard research aircraft and at observatory sites have allowed fingerprinting of different sources and quantification of timescales for plume processing by photochemistry and mixing, and coupled with atmospheric transport models these observations can now allow the detailed source history of a given air mass to be untangled.

RECOMMENDATION: Development of in situ techniques for measuring trace gas and aerosol species at higher temporal resolution, and with improved detection limits (e.g. <1pptv for NMHCs), would improve the detection of LRT events. Improved knowledge about NMHC emissions to better constrain estimates of chemical processing based on NMHC ratios is also required.

RECOMMENDATION: Assess the role of oxidants other than OH in pollutant processing, such as halogens and nitrate and the role of heterogeneous processes in polluted and non-polluted regions.

RECOMMENDATION: Further development of techniques based on the release and resampling of inert tracers to allow quantification of mixing processes during long-range transport.

2.4.3. Aerosol source attribution based on use of trace elements and isotopes

Although there now exist instrumental capabilities to measure the physical and chemical properties of pollutant aerosols in great detail, there are still relatively few reliable "tracers" that are uniquely source-specific; largely because aerosol source signatures (nature or anthropogenic) are rarely unique and because there is a large degree of atmospheric mixing during hemispheric-scale transport. In the case of natural particulate matter such as mineral dust, a combination of mineralogical and geochemical/isotopic properties can be used to identify the source regions(s). Over the past decade, a vast number of studies have documented the global dispersion of desert dust from East Asian deserts, in particular from northwestern China and southern Mongolia. Dust from these regions has been tracked as far as Alaska, the Yukon, Greenland and the European Alps [Bory et al., 2003; Grousset et al., 2003; Yasunari and Yamazaki, 2009; Zdanowicz et al., 2006]. Many of these studies make use of source-specific mineralogical abundances, elemental ratios, or the stable isotope ratios of strontium (Sr), eodymium (Nd) and lead (Pb) in the clay-size fraction of dust aerosols to identify their source(s). This has been facilitated by concerted efforts to map the compositional characteristics of different dust sources in eastern Asia [e.g. Yang et al., 2009; Zheng et al., 2009]. Trace element abundances and ratios also allow for trends in atmospheric mineral dust loading to be investigated in ice cores. Kaspari et al. [2009] recently developed a proxy record of dust aerosols from a Mt Everest ice core. The record suggests that Asian dust outbreaks have become more frequent since the 1800s, but provides no conclusive evidence of an anthropogenic impact on these trends. However, these trends are at odds with results from another core from the western Tibetan Plateau [Yang et al., 2006]. Meanwhile, a record from Mt Wrangell in Alaska suggests a recent increase in the trans-Pacific transport of Asian dust in the springtime [Yasunari et al., 2007].

For short-lived anthropogenic aerosols (e.g., trace metals, sulphate, and BC) the challenge of identifying distant sources is greater. This is because the compositional properties of these aerosols (e.g., elemental or isotopic abundances) are either not sufficiently characterized, or too variable, or both, to be unequivocally attributed to specific sources or source regions. Typically, compositional properties of pollutant aerosols are used to apportion total concentrations among various source types (e.g., fuel combustion vs. industrial refining) using some type of factor analysis, rather than to identify specific regional point sources [e.g. *Viana et al.*, 2008] although there are exceptions.

Lead (Pb) isotope ratios have proven particularly useful to trace back long-range airborne metal pollution to various source regions, because the stable Pb isotope composition of fumes from fuel combustion, or from some industrial processes, can carry a source-specific signature inherited from the ore from which the Pb was extracted [Komarek et al., 2008]. However, some pollution sources, such as base metal smelters, may have time-varying emission signatures [e.g. Simonetti et al., 2004]. More commonly, the Pb isotope ratios of aerosols are compared with the air mass signature of various possible source regions, and the relative contributions of these regions are estimated using a mass balance model such as the U.S. EPA's IsoSource [e.g. Sturges et al., 1993]. The essential requirement for this method is that the source signatures be well characterized. Fortunately, research efforts in recent decades have allowed the Pb isotope signatures of regional airsheds in both hemispheres to be defined [Bollhofer and Rosman, 2000; 2001; 2002]. Lead isotope measurements in aerosols and in snow and ice from Greenland, Arctic Canada and alpine regions have provided evidence of changing levels and sources of atmospheric Pb to the remote atmosphere over past decades and centuries [e.g. Mercier, 2000; Sherrell et al., 2000; Shotyk et al., 2005; Veyssevre et al., 2001]. These records show declining trace metal pollution after the 1980s in the North Atlantic sector and in western Europe, but a Pb isotope record from corals in the western Pacific testifies to a growing export of atmospheric Pb from China, which may have contributed up to 64 % of the Pb increase in the last two decades of the 20th century. This is in agreement with Pb isotope data from China [Mukai et al., 2001] and Pb deposition trends recorded in snow on the opposite side of the North Pacific [Osterberg et al., 2008]. A time series of aerosol Pb isotope data at a pair of sites on the west coast of California demonstrates how Pb isotope analysis has been used to resolve trans-Pacific versus local Pb sources [Ewing et al., 2010]. While the trans-Pacific Pb generally co-occurs with peaks in sample mass and silicon (Si) content due to trans-Pacific dust events, high Pb concentrations relative to sample mass and Si content are taken as evidence that the trans-Pacific Pb is primarily anthropogenic in origin (see Figure 2.22).

The source identification of anthropogenic sulphate aerosols is more challenging because there are also multiple natural sources that contribute to the observed sulphate burden in the atmosphere. However, advances in the stable isotope characterization of sulphur (δ^{34} S) in aerosols and precipitation now allow for the relative contributions of these source types to be estimated. As with Pb isotopes, the method requires that the δ^{34} S of possible reservoirs and sources (e.g., industrial SO₂ gases, marine biogenic sulphate) be determined with sufficient precision to discriminate among these sources. Also, sulphur isotope ratios in the atmosphere can be modified during heterogeneous phase reactions, in particular oxidation pathways, and the effects of these reactions must be quantified [Leung et al., 2001]. In pioneering studies, Nriagu et al. [1991] and Norman et al. [1999] used sulphur isotope measurements to estimate the relative contributions of natural (primarily marine) and nonmarine, primarily anthropogenic sources (including smelter emissions) to sulphate aerosols collected at Alert, Arctic Canada. Since then, several other studies have applied the method to characterize the sources of atmospheric sulphate in Europe [Novak et al., 2001], North America [Wadleigh et al., 2001], China [Mukai et al., 2001] and over the Atlantic Ocean [Patris et al., 2000; Wadleigh, 2004]. Measurements of δ^{34} S have also been applied to Greenland ice cores in order to document the changing burden of anthropogenic sulphate in the polar atmosphere since pre-industrial times [Patris et al., 2002]. As coal combustion remains one of the dominant man-made sources of SO₂ in the atmosphere, and as China emerges as the dominant emitter [Lu et al., 2010; Osterberg et al., 2008], efforts are now being directed at refining the sulphur isotope characterization of Chinese coal sources.



Figure 2.22. ²⁰⁸Pb/²⁰⁷Pb vs. ²⁰⁶Pb/²⁰⁷Pb in airborne particles at sites in California (Chabot Observatory, Mt. Tamalpais and central California surface sites), in San Francisco Bay waters, and from a variety of Chinese sources. Linear regression lines are shown in blue for wintertime Chabot samples, light brown for Chinese cities and loess, and dotted green for Mt. Tamalpais. [Reprinted from Figure 1 in Ewing, S. A., et al. (2010), Pb Isotopes as an Indicator of the Asian Contribution to Particulate Air Pollution in Urban California, *Environmental Science & Technology*, *44*(23): 8911-8916. Copyright 2010 American Chemical Society.]

Similarly, new analytical developments, in particular the measurement of carbon isotope ratios in organic aerosols, are now allowing for BC in air or deposited in snow to be apportioned among source types such as biomass burning, biofuel combustion, fossil fuel combustion, etc. [*Hegg et al.*, 2009]. The microcrystalline structure and trace element composition of soot may also reveal the type of emission process and help distinguish between, for example, BC emitted by industrial plants from that emitted by diesel engines [*VanderWal et al.*, 2010]. But the objective of fingerprinting specific regional sources of BC in long-range aerosols remains largely elusive.

With or without diagnostic tracers, attribution of aerosols to specific source regions can also be attempted by comparing observed temporal trends at receptor sites with emission trends from potential source regions. Ice cores from polar and alpine regions, which contain records of atmospheric aerosol deposition extending back over decades to millennia, have proven useful in this respect. While these records cannot, as yet, be directly interpreted in terms of aerosol loadings, the deposition histories developed from ice cores provide solid evidence of the intercontinental transport of anthropogenic aerosols from specific continental regions. In particular, the sources of atmospheric sulphate and/or nitrate aerosols have been investigated in Greenland [*Bigler et al.*, 2002], the Norwegian and Canadian Arctic [Goto-Azuma and Koerner, 2001; Kekonen et al., 2002; Moore et al., 2006], the European Alps [Preunkert et al., 2001; Preunkert et al., 2003] and western North America [Yalcin and Wake, 2001]. As well, the provenance of atmospheric Pb in the Arctic atmosphere has been investigated using cores from Greenland and the Canadian North [McConnell et al., 2002; Osterberg et al., 2008; Shotyk et al., 2005] (Figure 2.23). More recently, McConnell et al. [2007] developed a detailed record of BC deposition from central Greenland, which points to a predominance of North American sources prior to the mid-20th century, and the growing influence of east Asian sources since 1951.



Figure 2.23. Ice-core records of atmospheric lead (Pb) pollution in central Greenland (altitude ~3000 m) and Mt. Logan (altitude ~5500 m), Yukon Territory, Canada. Black lines are high-resolution measurements of Pb from Greenland [reprinted from Figure 1a in McConnell, J. R., et al. (2002), A 250-year high-resolution record of Pb flux and crustal enrichment in central Greenland, *Geophysical Research Letters, 29*(23): 2130-2133] and Mt. Logan [reprinted from Figure 3a in Osterberg, E., et al. (2008), Ice core record of rising lead pollution in the North Pacific atmosphere, *Geophysical Research Letters, 35*(L05810)]. Coloured symbols in upper panel are spot measurements from earlier studies. Also shown are reported industrial sulphur gas emissions from North America and Europe and from East Asia [from *Osterberg et al.*, 2008].

FINDING: Measurements of trace elements and isotopic ratios can provide useful fingerprints for different source types and emission regions influencing aerosol composition.

FINDING: Analysis of long-term aerosol and trace element records provides information about inter-annual variability in source attribution at a particular downwind measurement site as well as insights into how emissions may have changed in the past.

RECOMMENDATION: Further development is needed of isotope and geochemical fingerprinting techniques for the identification of different source types and, in the case of stable isotopes information about chemical processes occurring during transport. Proxy records of aerosol deposition (e.g. from ice cores) also provide useful tests for models over multi-decadal timescales.

2.4.4. Plume processing during long-range transport based on analysis of Lagrangian data and implications for global modelling of long-range pollutant transport

An ideal framework for assessing the processes influencing the evolution of pollutant concentrations during long-range transport away from source regions is to sample the same air mass several times at successively greater downwind distances. Such a Lagrangian approach assumes that an air mass can be tracked with sufficient accuracy during transport. Since this is, in reality, difficult to achieve, such attempts are often called pseudo-Lagrangian experiments. They were first attempted over rather short spatial (few hundred kilometres) and temporal scales (1-2 days) as part of, for example, the first and second Aerosol Characterization Experiments (ACE) [e.g. *Raes et al.*, 2000] or downwind of biomass burning regions over the southern Atlantic during TRACE-A [*Mauzerall et al.*, 1998].

Since the mid-2000s, as noted in Section 2.4.1, the ability to accurately forecast tracer (e.g., CO) distributions several days downwind from source regions has improved. This led to the planning and execution of the first inter-continental pseudo-Lagrangian experiment (IGAC Lagrangian 2K4), which took place as part of the ICARTT field campaign, to investigate long-range transport of

pollutants between North America and Europe. It was the first time that dedicated flights were planned to sample the same air masses on inter-continental scales with multiple aircraft based in North America, the Azores and Europe [*Fehsenfeld et al.*, 2006].

Several Lagrangian matches were identified post-campaign covering a variety of different meteorological situations [Methven et al., 2006]. One case is shown for illustration in Figure 2.24 when anthropogenic pollution was transported across the North Atlantic. Continuous, but slow, entrainment from the polluted low level outflow helped to maintain elevated pollutant levels (e.g., sulphate aerosol) in the marine boundary layer below. In an another case of pollution outflow, Real et al. [2008] used a photochemical model initialized with upwind data, and compared to downwind data allowing a detailed evaluation of the processes influencing O₃ levels during long-range transport. Initially O_3 and oxidized nitrogen concentrations were very high but hypothesized washout of HNO₃ combined with O₃ destruction was needed to explain the lower concentrations observed off the west coast of Ireland. Nevertheless, observed O₃ concentrations were still higher than average concentrations observed during summer at Mace Head, Ireland. This case also highlighted that high O₃ and water concentrations, observed close to the source regions, can lead to significant CO destruction making the use of CO as a tracer less than ideal in this case. In another case of long-range transport of an Alaskan forest fire plume, the trade-off between photochemical O₃ production from PAN decomposition and destruction due to increasing water vapour during descent into Europe were both found to be important as well as mixing with upper tropospheric air masses [*Real et al.*, 2007]. In both cases, it was difficult to determine an actual import of pollutants into the European PBL although in the latter case, the biomass burning plume was also observed in the lower troposphere in the south of France [Ravetta et al., 2007, see Section 2.2.1].



Figure 2.24. Trajectories 6 days backwards and forwards from the UK FAAM BAe146 flight track on 19 July 2004 (blue) showing links with upwind and downwind flights. Matching flights were: NASA DC8 15 July (dark green), NOAA WP-3D 15 July (light green), DLR Falcon 22 July (orange) and FAAM BAe146 25 July (red), numbered in time order. [Adapted from Figure 2b in Methven, J., et al. (2006), Establishing Lagrangian connections between observations within air masses crossing the Atlantic during the International Consortium for Atmospheric Research on Transport and Transformation, *Journal of Geophysical Research*, *111*(D23S62)] (case 1).

Analysis of the ICARTT forest fire case led to the application of parcel dispersion modelling techniques previously employed in the stratosphere to determine tropospheric mixing rates [*Pisso et al.*, 2009]. These approaches, based on the FLEXPART model and including a stochastic representation of mixing, were used to demonstrate the need for high spatial resolution calculations to reproduce observed plumes, many thousand kms downwind from source regions. Pisso et al. [2009] also estimated that global models need to be run with at least 40 km horizontal and 500 meter vertical

resolution in order to simulate long-range transport of pollutant plumes. Rastigejev et al. [2010] also showed that numerical diffusion, inherent in global Eulerian models, is dominating plume dilution even at increasing grid resolution. They suggest that adaptive gridding or embedded Lagrangian treatments of pollutant plumes are needed. Inclusion of photochemistry in the calculations of Pisso et al. [2009], representing resolutions of a few km, showed the importance of multiple air mass origins in governing measured concentrations of O₃ and CO along downwind flights [*Real et al.*, 2010]. An example is shown in Figure 2.25. Runs of the model reproduced observed correlation patterns and demonstrated the importance of chemical destruction as well as production in producing observed trace gas correlations. Results from a global model (MOCAGE) were unable to reproduce these correlations because the plume was too smeared out. Real et al.[2010] also estimated errors associated with resolution on net O₃ production showing that significant errors (up to 20-50%) can occur at plume edges compared to low resolution simulations which smear out gradients. Wild and Prather [2001] also quantified the impact of horizontal resolution on photochemical O₃ production, finding errors ranging from 27% at 5.5 degrees to 5% at 1.1 degrees. However, as already suggested errors might become non-linear below 1 degree.



Figure 2.25. O₃:CO correlations along Lagrangian plume match segment of a flight on 22 July 2004 by German DLR Falcon – measurements are shown as red diamonds; simulated results using a high resolution parcel model (ZooM-CiTTy) with chemistry are shown as blue rectangles; model runs without chemistry are shown as pink stars. [Adapted from Figure 10 in Real, E., et al. (2010), Toward a novel high-resolution modeling approach for the study of chemical evolution of pollutant plumes during long-range transport, *Journal of Geophysical Research 115*(D12302).]

FINDING: Pseudo-Lagrangian experiments provide unique and very useful constraints on the processing of long-range transport plumes by allowing the observation of air mass evolution in a flow-relative framework. Subsequent data analysis revealed detailed information regarding the photochemical evolution of anthropogenic and biomass burning plumes during long-range transport.

RECOMMENDATION: Lagrangian experiments should be used to rigorously assess the performance of global models, and the impact of resolution on long-range transport simulations requires further investigation. The comparison of model NMHC ratios with those observed would also provide constraints on models' abilities to represent the source contributions to long-range transport plumes, retain plume structure and representation of their dilution to the background.

FINDING: Developments in high-resolution Lagrangian modelling have allowed estimates of the resolutions needed to capture long-range transport of pollutant plumes in global models, which are currently too diffusive leading to the artificial dilution of pollutant plumes. The resolution issue is important for modelling pollutant events as well as modelling background pollutant levels where errors can occur if modelled lifetimes are incorrect. **RECOMMENDATION:** Improved horizontal and vertical resolution in global models together with the further development of plume in-grid techniques for pollutant plume transport is needed.

RECOMMENDATION: A better understanding of air mass entrainment from the free troposphere into the planetary boundary layer is required through dedicated field experiments, to improve estimates of the impacts of long-range ozone transport on regional air quality in receptor regions.

2.5. Research Needs

2.5.1. Surface Site Needs

A comprehensive ground-based global monitoring system is required for many purposes. including data provision for comparison with global models of chemistry and transport, and detection of trends in emissions of primary pollutants and their secondary products produced by atmospheric processing. The only truly global network for fulfilling this requirement is contained in the World Meteorological Organization (WMO) Global Atmospheric Watch (GAW) program which organizes the measurements of a large range of chemical species at sites distributed globally (http://www.wmo.int/pages/prog/arep/gaw/gaw home en.html). The data are stored at the World Data Centre for Greenhouse Gases (WDCGG). Figure 2.26 shows the locations of the sites for measurement of reactive and greenhouse gases. This global network is complemented by national and transnational networks that cover regions e.g. EMEP in Europe, the Clean Air Status and Trends Network (CASTNET) in the US, and the Canadian Air and Precipitation Monitoring Network (CAPMoN). For the detection of episodes of long-range intercontinental transport of gaseous pollutants however, only a few sites are capable of producing useful information. For aerosols it is a different matter and many sites in remote locations reveal transport events. As discussed in Section 2.2.2, mountaintop sites are particularly useful for detecting long-range transport events arriving on the western coasts of North America and Europe and even Europe to Asia transport.



Figure 2.26. Network of surface-based remote observatories organized through the World Meteorological Organization's Global Atmospheric Watch (GAW) Program.

The WMO GAW program measures gaseous species, including CO, O_3 , selected volatile organic compounds (VOCs), NO_x and NO_y compounds, and SO_2 . A new global network for measuring VOCs has just been created within the GAW program and a network for global measurements of NO_x and NO_y compounds is planned. Unfortunately, no global network exists for SO_2 although this species, and all the others mentioned, are measured by EMEP and CASTNET.

These regional networks were set up to follow trends in emissions of primary pollutants and to establish the regional background of secondary pollutants which can be strongly influenced by long-range transport from region to region. While some networks exist in southeast Asia, such as in Taiwan and Japan, there is a need to further develop long-term measurement networks in this important emission region, and especially in China and other parts of southeast Asia.

RECOMMENDATION: Continue current ground-based networks into the future with additions made to the range of constituents measured and additional sites in optimal locations for characterization of long-range transport and long-term changes in pollutant concentrations. Such networks require continued maintenance with proper attention paid to calibration and data quality issues.

2.5.2. Vertical Profiling Needs

Instrumented surface sites provide continuous in-situ measurements of trace gases and aerosols that are valuable for identifying annual trends as well as seasonal and diurnal variability. However, with the exception of a few high mountain top sites, they are only representative of the lowest few hundred meters of the atmosphere and need to be supplemented with vertical profiling through the full troposphere to allow, in particular better quantification of the export and import of pollutants to and from emission regions. This requires frequent vertical profiling at key locations on the upwind and downwind edges of the continents and especially in regions susceptible to pollution import (see Chapter 1) where long-range transport of pollutants may lead to exceedence of pollution thresholds for air quality and crops. The three methods currently used for the vertical profiling of O_3 and PM are described below.

Ozonesondes

These balloon-borne instruments are routinely launched year-round at approximately 35 sites in the northern hemisphere, typically one to three times per week, providing measurements at 100 m resolution through the troposphere. However very few sites are ideally located for determining the baseline O₃impacting a continent (Figure 2.27). While ozonesonde datasets have provided information about long-term changes in O₃ concentrations [e.g. *Oltmans et al.*, 2006], difficulties arise associated with changes in techniques over time and calibration, which make interpretation of these records problematic [*Schnadt Poberaj et al.*, 2009; *Smit et al.*, 2007]. A recent study reported increasing free tropospheric O₃ over western North America during springtime, 1995-2008, using large data sets composed primarily of MOZAIC data (see below). This study found that weekly ozonesonde profiles at a particular location were not frequent enough to detect increasing O₃ trends in the free troposphere [*Cooper*, 2010]. Detection of the trend would require ozonesonde profiles on a daily basis. It can be noted that the annual cost of operating a hypothetical northern hemisphere ozonesonde network of 60 sites with daily profiles is on the order of 20 million US\$.

Ozone and aerosol lidars

Lidars are ground-based lasers that measure O_3 mixing ratios and PM loadings with vertical resolution better than 1 km through much of the troposphere, with some having the capability to reach the tropopause. These instruments cannot however measure through clouds. Figure 2.27 shows the locations of most of the tropospheric ozone and aerosol lidars in the northern hemisphere. There are very few O_3 lidars, all in North America, Europe and New Zealand, while the number of aerosol lidars is at least 70 with a large proportion in East Asia. As noted in Sections 2.2 and 2.3, ground-based lidars contribute very valuable information about long-range transport of O_3 and PM.

Commercial aircraft

Since 1994 the European MOZAIC program (http://mozaic.aero.obs-mip.fr) has measured O_3 , water vapour, CO and NO_y from up to five commercial Airbus aircraft. To date the program has measured over 60,000 vertical profiles, mostly in Europe, the eastern U.S. and Japan. The most frequently sampled cities are shown in Figure 2.27. The data have been used for a wide variety of studies and, as shown in the recent study by Cooper et al. [2010], can be used for the analysis of O_3 trends in the free troposphere. Some limitations of the program are that measurements in the



Figure 2.27. Current locations in the Northern Hemisphere with frequent ozone and PM profiling of the troposphere by: aerosol lidars (green +) [*Bosenberg and Hoff*, 2008], ozone lidars (yellow x) and weekly ozonesondes (white/red dots) (www.woudc.org). Also shown are the cities frequently sampled by MOZAIC/IAGOS commercial aircraft (magenta x) during 1995-2010.

lowermost troposphere are contaminated by local airport effects, data are collected from a finite number of cities, flight routes are often biased to certain times of day, and scientists have no control over where profiles occur. The MOZAIC program is being replaced by the new IAGOS program (http://www.iagos.org/) from mid-2010 with an increased number of measurements of trace gases (O₃, CO, CO₂, NO_y, NO_x, and water), PM and cloud particles. The goal is an initial fleet of 10-20 long-range commercial aircraft operated by a range of international carriers providing cost-effective profiling (approximately \$500 per profile) around the world. The data will be available in near real time to weather services and Earth observation service centres. In addition, the CARIBIC (Civil Aircraft for Regular Investigation of the Atmosphere Based on an Instrument Container) [*Brenninkmeijer et al.*, 2007] program that began in 1997 has joined IAGOS. The latest phase of CARIBIC uses one commercial A340 Airbus conducting approximately 40 flights per year between Frankfurt and destinations around the world. While the CARIBIC aircraft flies less frequently than its MOZAIC counterparts, it makes a wider range of measurements, which includes O₃ and PM (particle number, size distribution and composition).

Routine monitoring from dedicated aircraft could also augment future vertical profiling of O₃ and PM. For example, the NOAA Earth System Research Laboratory contracts privately owned aircraft to make routine vertical measurements of carbon cycle gases from sixteen locations across North America. A similar program could be created to measure O₃ and PM. The recent development of Un-manned Aerial Vehicles (UAVs) that can carry significant payloads (up to 900 kg), fly up to 30 hours and reach altitudes near 20km (see e.g., http://uas.noaa.gov/ and http://www.nasa.gov/centers/dryden/aircraft/GlobalHawk/), could eventually provide the ideal monitoring for quantifying long-range transport of O₃ and PM in the northern hemisphere. Such aircraft could be flown several times per week on routine survey missions along the western coast of a receptor region. However, the cost of such a program would be large, and the use of UAVs in the capacity of routine air quality monitoring has not yet been fully developed.

FINDING: Lidars, ozonesondes and commercial aircraft provide frequent profiling of O₃ and PM at many locations across the northern hemisphere. However, very few of the profiling locations are ideally situated to quantify baseline O₃ and PM flowing into the continents.

RECOMMENDATION: Additional profile measurements are much needed to provide information on the vertical distribution of pollutant layers Commercial aircraft measurements should be expanded, especially within the continents (North America, Asia, Europe), and with additional measurements of trace gases and aerosols. Additional ground-based lidars should be sited to yield the best quantification of baseline O₃ and PM impacting downwind continents.

RECOMMENDATION: Development is needed of the capability to use unmanned aerial vehicles for measuring pollutant profiles in regions of pollutant import over receptor regions.

RECOMMENDATION: Maintenance of the long-term record from ozonesondes is required.

RECOMMENDATION: Added emphasis should be placed upon measurements designed to quantify the import of pollutants into the planetary boundary layer of receptor continents.

2.5.3. Satellite Data Needs

Satellite remote sensing provides a valuable data source to assess the impacts of long-range transport and derive pollutant emissions. Efforts in a few key areas offer high potential to further enhance this capability. One such area is data access where excellent examples exist of publicly available satellite data, often in near real-time. Making all scientific satellite data available for free in an easily accessible and timely manner would increase the value of the measurements to the wider scientific community (e.g. through tools like Giovanni (http://disc.sci.gsfc.nasa.gov/giovanni), and TEMIS (www.temis.nl)). This should include regular updates from data providers following product improvements together with documentation detailing, in particular, the quality of the data and methodology for its scientific use.

Further quantification of error estimates and improvement in satellite retrievals are also needed. Great progress has been made in developing retrieval algorithms that yield quantitative results that can be compared with models and other measurements in a consistent manner. Nonetheless, continued effort is needed to achieve a clear understanding of the accuracy and precision of these measurements. In addition to an assessment and quantification of the main error sources in the retrieval processes under different characteristic conditions (background, urban pollution, biomass burning, etc.) and measurement scenarios (dependent on meteorology, surface type, time of day etc.) systematic comparisons against independent measurements are necessary. A challenge is that surface and aircraft measurements available for retrieval evaluation are often at different spatial and temporal scales than the satellite measurements, and have different inherent measurements sensitivities to the atmospheric quantity of interest, which can lead to ambiguous comparisons. Therefore, although the development of systematic vertical profiling (Section 2.5.2) would clearly help, dedicated validation activities are needed that span a satellite pixel (typically ~15km for recent instruments such as IASI/METOP), and include profile information, for a range of pollutants, seasons and atmospheric conditions. This evaluation effort must persist over the entire mission duration to establish confidence in trend detection in the presence of possible instrumentation drifts over time.

Continued investment in retrieval development could contribute to significant advances in assessment of long-range transport. For example, nascent tropospheric HNO₃ retrievals [*Wespes et al.*, 2009] could provide insight into O₃ sources. Emerging developments in retrievals of ammonia [*Beer*, 2008; *Clarisse et al.*, 2009] and SO₂ [*Lee et al.*, 2009] offer a constraint on PM sources. The development of multispectral retrieval techniques and better instrumental spectral resolution have the potential to improve vertical profile information and allow discrimination of the lowermost troposphere from the free troposphere for species such as CO [*Deeter et al.*, 2009] and O₃ [*Liu et al.*, 2010; *Worden et al.*, 2007]. In addition to these passive observations, innovative approaches using active remote sensing (the CALIOP lidar in space onboard CALIPSO since 2006 and the future ATLID onboard the EarthCare mission) provide valuable complementary information on the altitude and shape of the transported plumes.

Finally, most current satellite missions able to make measurements of processes relevant for long-range transport are maturing could soon end. For example, with the exception of GOME-2 and IASI-METOP, the instruments in Table 2.1 are all operating beyond their planned lifetimes. Planning for the next generation of satellites is beginning, but progress is slow due to limited resources, and the current wealth of data is unlikely to be maintained over the next few years as the current instruments expire. Proposals for future missions are recognizing the complementary nature of measurements of trace gases and PM from both low Earth orbit (LEO), which provides global coverage, and geostationary orbit (GEO), which allows for continental-scale measurements at relatively high spatiotemporal resolution. Indeed, a tantalizing possibility for the future would be the international implementation of a constellation of LEO and GEO platforms providing detailed coverage of most of

the globe. This would permit comprehensive investigation of pollution events, transport and atmospheric composition processes at different altitudes and across spatial and temporal scales.

FINDING: Current satellite datasets for trace gases and aerosols provide critical spatial and temporal information for improved understanding of transport pathways and emission strengths.

FINDING: Many current satellites measuring atmospheric composition are reaching the end of their missions so future capabilities may be reduced.

RECOMMENDATION: Continued efforts are required to improve retrieval accuracy, to improve vertical resolution, and to develop modelling tools to interpret the observations.

RECOMMENDATION: The development and launch of geostationary satellites measuring trace gas and aerosols is needed provide much improved temporal coverage over key emission regions of the northern hemisphere.

RECOMMENDATION: High priority should be given to developing future low Earth orbit missions, accelerating the implementation timetable, and fostering international cooperation on shared use of available data and coordinated future mission planning.

2.5.4. Intensive Campaign Needs

Intensive field campaigns provide abundant opportunities to advance our understanding of the hemispheric transport of air pollution. Each campaign generally deploys a wide array of newly developed, state-of-the-art instruments in a coordinated manner on several mobile (aircraft, ship) platforms and at surface sites. These instruments include in-situ and remote-sensing measurements. Strategies to answer open research questions guide the planning of such studies, so the results invariably advance our understanding on multiple fronts.

Issues relevant to long-range pollutant transport that can be addressed by intensive campaigns using instrumented aircraft are manifold. Emission sources can be characterized and their emissions quantified for primary pollutants, greenhouse gases, and precursors of O_3 and PM. The chemical transformation of emitted species can be followed through the formation of secondary species (both gas-phase and aerosol), and on to the ultimate destruction and deposition processes. Atmospheric transport mechanisms can be identified and their relative importance determined. This includes not only transport of pollutants away from emission regions but also their long-range transport to receptor regions and import into the boundary from the free troposphere. This latter aspect has yet to be investigated in detail in many regions.

In addition to addressing specific issues, intensive field campaigns provide extensive and detailed data sets that serve as benchmarks for testing chemical-transport model simulations. In particular, Lagrangian experiments making multiple samplings of the same air mass during long-range transport between continents can provide important information on chemical and aerosol processing as well as deposition processes and mixing rates in the troposphere.

Although far less expensive than a single satellite instrument, an intensive campaign does challenge the resources that are typically available for field measurements at a national level. Hence, the larger campaigns are beyond the capabilities of a single agency or even nation, and therefore must be planned wisely and coordinated at an international level.

FINDING: Intensive field campaigns provide one of our most important means of advancing our understanding of atmospheric chemistry and transport in general, and the long-range transport of pollutants in particular. Lagrangian experiments are particularly useful in this regard.

RECOMMENDATION: Intensive field campaigns should be conducted frequently focusing on key issues where large uncertainties remain in our understanding of long-range transport of pollutants between continents such as the import of pollutants into the boundary layer over downwind receptor regions. Given the extensive resources required, it is essential to carefully plan such experiments, ideally within international frameworks that can mobilize a large scientific community to address a particular issue.

RECOMMENDATION: Continued development of high quality, accurate measurements of trace gases and aerosols, in conjunction with quality control assessments, for use in intensive field campaigns is needed. The development of methods based on, for example, the release of inert tracers and fingerprinting techniques (using in-situ measurements of isotopes, VOCs), would allow significant advances in our ability to follow pollution plumes, and in source identification and verification of emission inventories.

2.5.5. Model evaluation against observations

The results presented in this chapter have focused on observational evidence for long-range transport of pollutants. Data analyses using multiple parcel models or trace gas correlations/ratios were also presented as a means of identifying source regions. Global CTMs are the tools used to estimate the impact of different emission regions on downwind regions (see Chapter 4). Here we comment on their usefulness for this purpose and the methodologies employed for evaluating their performance in terms of ability to simulate pollutant long-range transport. Comparison with observations is generally used as a means of evaluating model performance. Previous studies analyzing multi-model ensembles [e.g. Dentener et al., 2005; Fiore et al., 2009] largely focused on comparison with surface data. For example, Fiore et al. [2009] attempted to assess model skill by comparing with surface O_3 observations binned over different sub-regions. While certain biases were evident, it was difficult to attribute causes, in part due the large differences in, for example modelled VOC emissions, which varied by a factor of 10 across models. Other studies have compared models with monthly mean ozonesonde data [e.g., Stevenson et al., 2006]. Such comparisons do not test model ability to simulate observed variability including particular episodes. Accurately reproducing such data and attributing sources remains a challenge for current global models. Additional development and application of computationally efficient algorithms (such as adjoint approaches) are valuable to calculate model sensitivities to multiple parameters, such as long-range transport or emission sources.

Detailed comparison of specific global models with data collected as part of intensive field campaigns does allow advances in our understanding of processes influencing the model results and discrepancies with the data. For example, Hudman et al. [2008] used INTEX-NA data to conclude that EPA CO emissions are too high by 60%. Modelling associated with analysis of Lagrangian data also provides important information about the processes governing pollutant concentrations during particular long-range transport events (see Section 2.4.4). Results showed that plume signatures are diluted too rapidly in current global models due to too coarse resolution [*Pisso et al.*, 2009; *Real et al.*, 2010] and diffusive advection schemes [*Rastigejev et al.*, 2010] confirming the need to run global Eulerian models at higher resolution or to include in-grid plume treatments.

Many datasets exist which can be further exploited to improve model capabilities in terms of long-range pollutant transport. In particular, this includes satellite data and vertical profile data from, for example, the MOZAIC program. The ability of models to reproduce the full spectrum of measured concentrations (e.g. Figure 2.1), measured long-term trends (e.g. Figures 2.7 and 2.8), and the relationships among measured species needs to be assessed. The continued development of a dedicated data portal to house or link to relevant datasets would be beneficial in this regard.

FINDING: Current datasets (surface, lidar, ship, aircraft, sonde and satellite) provide information about monthly, seasonal and inter-annual variability of certain trace gases and aerosol species, which can be used to assess model performance.

FINDING: Multi-species analysis of field campaign data often provides new insights into chemical and aerosol processing leading to improvements in their representation in CTMs.

RECOMMENDATION: Increased use of vertical profile data is needed, especially data collected at high temporal resolution (daily) and satellite data for the evaluation of global model performance and for quantifying the impact of emissions from one region on downwind regions.

RECOMMENDATION: Combined use of campaign and routine monitoring data to assess and improve model treatment of pollutant import into the lower troposphere over downwind receptor regions is needed.

References

- Andreae, M. O., et al. (1988), Vertical distribution of dimethylsulfide, sulfur dioxide, aerosol ions, and radon over the Northeast Pacific Ocean, *Journal of Atmospheric Chemistry*, 6(1): 149-173.
- Arnold, S. R., et al. (2007), Statistical inference of OH concentrations and air mass dilution rates from successive observations of nonmethane hydrocarbons in single air masses, *Journal of Geophysical Research*, 112(D10S40).
- Barrie, L. A., et al. (2001), A comparison of large scale atmospheric sulphate aerosol models (COSAM): overview and highlights, *Tellus Series B Chemical and Physical Meteorology*, 53(5): 615-645.
- Beer, R., et al. (2008), First satellite observations of lower tropospheric ammonia and methanol, *Geophysical Research Letters*, *35*(L09801).
- Ben-Ami, Y., et al. (2009), Patterns of North African dust transport over the Atlantic: winter vs. summer, based on Calipso first year data, *Atmospheric Chemistry and Physics*, 9: 7867-7875.
- Bertschi, I. T., et al. (2004), PHOBEA/ITCT 2002 airborne observations of transpacific transport of ozone, CO, volatile organic compounds, and aerosols to the northeast Pacific: Impacts of Asian anthropogenic and Siberian boreal fire emissions, *Journal of Geophysical Research*, *109*(D23S12).
- Bigler, M., et al. (2002), Sulphate record from a northeast Greenland ice core over the last 1200 years based on continuous flow analysis, *Annals of Glaciology*, *35*: 250-256.
- Bodhaine, B. A., and E. G. Dutton (1993), A long-term decrease in Arctic haze at Barrow, Alaska, *Geophysical Research Letters*, 20(10): 947-950.
- Bollhofer, A., and K. Rosman, Jr. (2000), Isotopic source signatures for atmospheric lead: The Northern Hemisphere, *Geochimica et Cosmochimica Acta*, 64: 3251-3262.
- Bollhofer, A., and K. Rosman, Jr. (2001), Isotopic source signatures for atmospheric lead: The Southern Hemisphere, *Geochimica et Cosmochimica Acta*, 65: 1727-1740.
- Bollhofer, A., and K. Rosman, Jr. (2002), The temporal stability in lead isotopic signatures at selected sites in the Southern and Northern Hemispheres, *Geochimica et Cosmochimica Acta*, 66: 1375-1386.
- Bonasoni, P., et al. (2008), The ABC-Pyramid Atmospheric Observatory in Himalaya for aerosol, ozone and halocarbon measurements, *Science of the Total Environment*, *391*: 252-261.
- Bory, A. J. M., et al. (2003), Two distinct seasonal Asian source regions for mineral dust deposited in Greenland (NorthGRIP), *Geophysical Research Letters*, *30*(4): 1167-1170.
- Bosenberg, J., and R. Hoff (2008), Plan for the implementation of the GAW Aerosol Lidar Observation Network GALION, 52 pp, World Meteorological Organization, Hamburg, Germany. *WMO TD No.* 1443
- Brenninkmeijer, C. A. M., et al. (2007), Civil aircraft for the regular investigation of the atmosphere based on an instrumented container: The new CARIBIC system, *Atmospheric Chemistry and Physics*, 7: 5277-5339.
- Brock, C. A., et al. (2004), Particle characteristics following cloud-modified transport from Asia to North America, *Journal of Geophysical Research*, 109(D23S26).
- Bronnimann, S., et al. (2002), Trends in near surface ozone concentrations in Switzerland, *Atmospheric Environment*, *36*: 2841-2853.
- Chan, C. Y., et al. (2003), Urban and background ozone trend in 1984-1999 at subtropical Hong Kong, South China, *Ozone: Science & Engineering*, 25(6): 513-522.

- Chin, M., et al. (1994), Relationship of ozone and carbon monoxide over North America, *Journal of Geophysical Research*, 99(D7): 14565-14574.
- Chou, C. C.-K., et al. (2006), The trend of surface ozone in Taipei, Taiwan, and its causes: Implications for ozone control strategies, *Atmospheric Environment*, 40(21): 3898-3908.
- Clarisse, L., et al. (2009), Global ammonia distribution derived from infrared satellite observations, *Nature Geoscience*, *2*(7): 479-483.
- Cooper, O. R., et al. (2004), A case study of trans-Pacific warm conveyor belt transport: The influence of merging airstreams on trace gas import to North America, *Journal of Geophysical Research*, 109(D23S08).
- Cooper, O. R., et al. (2010), Increasing springtime ozone mixing ratios in the free troposphere over western North America, *Nature* 463: 344-348.
- Creilson, J. K., et al. (2003), Intercontinental transport of tropospheric ozone: a study of its seasonal variability across the North Atlantic utilizing tropospheric ozone residuals and its relationship to the North Atlantic Oscillation, *Atmospheric Chemistry and Physics*, 3: 2053-2066.
- Crutzen, P. J. (1988), Tropospheric ozone: an overview, in *Tropospheric Ozone Regional and Global Scale Interactions*, edited by I. S. A. Isaksen, 3-33 pp., D. Reidel, Dordrecht.
- Damoah, R., et al. (2004), Around the world in 17 days hemispheric-scale transport of forest fire smoke from Russia in May 2003, *Atmospheric Chemistry and Physics*, 4: 1311-1321.
- Deeter, M. N., et al. (2009), CO retrievals based on MOPITT near-infrared observations, *Journal of Geophysical Research*, 114(D04303).
- Dentener, F., et al. (2005), The impact of air pollutant and methane emission controls on tropospheric ozone and radiative forcing: CTM calculations for the period 1990-2030, *Atmospheric Chemistry and Physics*, 7(5): 1731-1755.
- Derwent, R. G., et al. (1997), Observation and interpretation of the seasonal cycles in the surface concentrations of ozone and carbon monoxide at Mace Head, Ireland, from 1990 to 1994, *Atmospheric Environment*, *32*(2): 145-157.
- Derwent, R. G., et al. (2007), Trends over a 20-year period from 1987 to 2007 in surface ozone at the atmospheric research station, Mace Head, Ireland, *Atmospheric Environment*, 41(39): 9091-9098.
- Ding, A., et al. (2009), Transport of north China air pollution by midlatitude cyclones: Case study of aircraft measurements in summer 2007, *Journal of Geophysical Research*, *114*(D08304).
- Ding, A. J., et al. (2008), Tropospheric ozone climatology over Beijing: Analysis of aircraft data from the MOZAIC program, *Atmospheric Chemistry and Physics*, 8: 1-13.
- Dubovik, O., et al. (2002), Variability of absorption and optical properties of key aerosol types observed in worldwide locations, *Journal of Atmospheric Science*, *59*: 590-608.
- Duce, R. A., et al. (1980), Long-range atmospheric transport of soil dust from Asia to the Tropical North Pacific: Temporal variability, *Science*, 209: 1522-1524.
- EARLINET (2003), EARLINET: A European Aerosol Research Lidar Network to establish an Aerosol Climatology, Final Report for the Period February 2000 to February 2003, Max Planck Institut fur Meteorologie, Hamburg, Germany.
- Edwards, D. P. (2006), Air quality remote sensing from space, *Eos, Transactions of the American Geophysical Union*, 87(33).
- Elguindi, N., et al. (2010), Current status of the ability of the GEMS/MACC models to reproduce the tropospheric CO vertical distribution as measured by MOZAIC, *Atmospheric Chemistry and Physics Discussions*, *3*: 391-449.
- EMEP (2004), EMEP Assessment, Part I: European Perspective, edited by G. Lovblad, et al., 180 pp., European Monitoring and Evaluation Programme, Oslo, Norway.

- EU (2006), Remote Sensing of Air Pollution, edited by A. Borowiak and F. Dentener, 64 pp., Institute for Environment and Sustainability 2006, European Commission, Ispra, Italy. EUR 22330 EN.
- Ewing, S. A., et al. (2010), Pb Isotopes as an Indicator of the Asian Contribution to Particulate Air Pollution in Urban California, *Environmental Science & Technology*, 44(23): 8911-8916.
- Fehsenfeld, F. C., et al. (2006), International Consortium for Atmospheric Research on Transport and Transformation (ICARTT): North America to Europe—Overview of the 2004 summer field study, *Journal of Geophysical Research*, 111(D23S01).
- Feister, W., and W. Warmbt (1987), Long-term measurements of surface ozone in the German Democratic Republic, *Journal of Atmospheric Chemistry*, 5: 1-21.
- Fiore, A. M., et al. (2009), Multimodel estimates of intercontinental source-receptor relationships for ozone pollution, *Journal of Geophysical Research*, *114*(D04301).
- Fischer, E. V., et al. (2009), Meteorological Controls on Observed Peroxyacetyl Nitrate (PAN) at Mount Bachelor during the spring of 2008, *Journal of Geophysical Research*, *115*(D03302).
- Fishman, J., et al. (2008), Remote sensing of tropospheric pollution from space, *Bulletin of the American Meteorological Society*, 89(6): 805-821.
- Forster, C., et al. (2001), Transport of boreal forest fire emissions from Canada to Europe, *Journal of Geophysical Research*, *106*(D19): 22887-22906.
- Fuhrer, J., et al. (1997), Critical levels for ozone effects on vegetation in Europe *Environmental Pollution*, 97(1-2): 91-106.
- Gerbig, C., et al. (2003), Toward constraining regional-scale fluxes of CO2 with atmospheric observations over a continent: 2. Analysis of COBRA data using a receptor-oriented framework, *Journal of Geophysical Research*, 108(D244757).
- Goto-Azuma, K., and R. Koerner (2001), Ice-core studies of anthropogenic sulfate and nitrate trends in the Arctic, *Journal of Geophysical Research*, *106*(D5): 4959-4969.
- Grousset, F. E., et al. (2003), Case study of a Chinese dust plume reaching the French Alps, *Geophysical Research Letters*, *30*(6).
- Guerova, G., et al. (2006), Impact of transatlantic transport episodes on summertime ozone in Europe, *Atmospheric Chemistry and Physics*, 6: 2057-2072.
- Hamelin, B., et al. (1989), Lead isotopes in trade wind aerosols at Barbados: The influence of European emissions over the North Atlantic, *Journal of Geophysical Research*, 94(C11): 16243–16250.
- Heald, C. L., et al. (2003), Asian outflow and trans-Pacific transport of carbon monoxide and ozone pollution: An integrated satellite, aircraft, and model perspective, *Journal of Geophysical Research*, 108(D244804).
- Heald, C. L., et al. (2006), Transpacific transport of Asian anthropogenic aerosols and its impact on surface air quality in the United States, *Journal of Geophysical Research*, 111(D14310).
- Hegg, D. A., et al. (2009), Source attribution of Black Carbon in Arctic snow, *Environmental Science & Technology*, 43: 4016-4021.
- Heidam, N. Z., et al. (2004), Arctic atmospheric contaminants in NE Greenland: levels, variations, origins, transport, transformations and trends 1990-2001, *Science of The Total Environment*, 331(1-3): 5-28.
- Helmig, D., et al. (2008), Nonmethane hydrocarbons at Pico Mountain, Azores: 1. Oxidation chemistry in the North Atlantic region, *Journal of Geophysical Research*, *113*(D20S91).
- Henze, D. K., et al. (2009), Inverse modeling and mapping US air quality influences of inorganic PM_{2.5} precursor emissions using the adjoint of GEOS-Chem, *Atmospheric Chemistry and Physics*, 9: 5877-5903.
- Hirdman, D., et al. (2010), Source identification of short-lived air pollutants in the Arctic using statistical analysis of measurement data and particle dispersion model output, *Atmospheric Chemistry and Physics*, *10*: 669-693.

- Honrath, R. E., et al. (2004), Regional and hemispheric impacts of anthropogenic and biomass burning emissions on summertime CO and O3 in the North Atlantic lower free troposphere, *Journal of Geophysical Research*, 109(D24310).
- Honrath, R. E., et al. (2008), Nonmethane hydrocarbons at Pico Mountain, Azores: 2. Event-specific analysis of the impacts of mixing and photochemistry on hydrocarbon ratios, *Journal of Geophysical Research*, 113(D20S92).
- Horowitz, L. W. (2006), Past, present, and future concentrations of tropospheric ozone aerosols: Methodology, ozone evaluation, and sensitivity to aerosol wet removal, *Journal of Geophysical Research*, *111*(D22211).
- Hudman, R. C., et al. (2007), Surface and lightning sources of nitrogen oxides over the United States: Magnitudes, chemical evolution, and outflow, *Journal of Geophysical Research*, *112*(D12S05).
- Hudman, R. C., et al. (2008), Biogenic versus anthropogenic sources of CO in the United States, *Geophysical Research Letters*, 35(L04801).
- Huntrieser, H., et al. (2005), Intercontinental air pollution transport from North America to Europe: Experimental evidence from airborne measurements and surface observations, *Journal of Geophysical Research*, *110*(D01305).
- Igarashi, Y., et al. (2006), Seasonal variations in SO2 plume transport over Japan: Observations at the summit of Mt. Fuji from winter to summer, *Atmospheric Environment*, 40(36): 7018-7033.
- Jaffe, D., et al. (2003), The 2001 Asian Dust Events: Transport and Impact on Surface Aerosol Concentrations in the U.S., *Eos, Transactions American Geophysical Union*, 84(46): 501-507.
- Jaffe, D., et al. (2005a), Seasonal cycle and composition of background fine particles along the west coast of the US *Atmospheric Environment*, *39*(2): 297-306.
- Jaffe, D., and J. Ray (2007), Increase in surface ozone at rural sites in the western US, *Atmospheric Environment*, 41(26): 5452-5463.
- Jaffe, D. A., et al. (2004), Long-range transport of Siberian biomass burning emissions and impact on surface ozone in western North America, *Geophysical Research Letters*, *31*(L16106).
- Jaffe, D. A., et al. (2005b), Export of atmospheric mercury from Asia, *Atmospheric Environment*, 39(17): 3029-3038.
- Kalashnikova, O. V., and R. A. Kahn (2008), Mineral dust plume evolution over the Atlantic from MISR and MODIS aerosol retrievals, *Journal of Geophysical Research*, *113*(D24204).
- Karnieli, A., et al. (2009), Temporal trend in anthropogenic sulfur aerosol transport from central and eastern Europe to Israel, *Journal of Geophysical Research*, *114*(D00D19).
- Kaspari, S., et al. (2009), A high-resolution record of atmospheric dust composition and variability since A.D. 1650 from a Mount Everest ice core, *Journal of Climate*, 22(14).
- Kaufman, Y. J., et al. (2005), Dust transport and deposition observed from the Terra-Moderate Resolution Imaging Spectroradiometer (MODIS) spacecraft over the Atlantic Ocean, *Journal of Geophysical Research*, 110(D10S12).
- Keating, T., et al. (2005), Air quality impacts of intercontinental transport, *EM / Air and Waste Management Association*(October): 28-30.
- Kekonen, T., et al. (2002), An 800 year record of nitrate from the Lomonosovfonna ice core, Svalbard, *Annals of Glaciology*, 35: 261-265.
- Koch, D., et al. (2009), Evaluation of black carbon estimations in global aerosol models, *Atmospheric Chemistry* and Physics, 9: 9001-9026.
- Komarek, M., et al. (2008), Lead isotopes in environmental sciences: A review, *Environment International*, 34: 562-577.
- Lamarque, J.-F., et al. (2005), Tropospheric ozone evolution between 1890 and 1990, *Journal of Geophysical Research*, *110*(D08304).

- Lamarque, J. F., et al. (2010), Historical (1850-2000) gridded anthropogenic and biomass burning emissions of reactive gases and aerosols: Methodology and application, *Atmospheric Chemistry and Physics*, 10: 7017-7039.
- Lee, C., et al. (2009), Retrieval of vertical columns of sulfur dioxide from SCIAMACHY and OMI: Air mass factor algorithm development and validation, *Journal of Geophysical Research*, *114*(D22303).
- Legras, B., et al. (2003), Vertical diffusivity in the lower stratosphere from Lagrangian back-trajectory reconstructions of ozone profiles, *Journal of Geophysical Research*, *108*(D18): 4562-4570.
- Leung, F.-Y., et al. (2001), Sulfur isotopic fractionation in the gas-phase oxidation of sulfur dioxide initiated by hydroxyl radicals, *Journal of Physical Chemistry*, 105: 8073-8076.
- Lewis, A. C., et al. (2007), Chemical composition observed over the mid-Atlantic and the detection of pollution signatures far from source regions, *Journal of Geophysical Research*, *112*(D10S39).
- Li, Q., et al. (2004), Export of NO_y from the North American boundary layer: Reconciling aircraft observations and global model budgets, *Journal of Geophysical Research*, *109*(D02313).
- Li, S.-M., and L. A. Barrie (1993), Biogenic Sulfur Aerosol in the Arctic Troposphere: 1. Contributions to Total Sulfate, *Journal of Geophysical Research*, *98*(D11): 20613-20622.
- Li, Z., et al. (2009), Uncertainties in satellite remote sensing of aerosols and impact on monitoring its long-term trend: A review and perspective, *Annales Geophysicae*, 27: 2755-2770.
- Liang, Q., et al. (2007), Summertime influence of Asian pollution in the free troposphere over North America, *Journal of Geophysical Research*, *112*(D12S11).
- Likens, G. E., and F. H. Bormann (1974), Acid rain: a serious regional environmental problem, *Science*, *184*(4142): 1176-1179.
- Lin, J. C., et al. (2003), A near-field tool for simulating the upstream influence of atmospheric observations: The Stochastic Time-Inverted Lagrangian Transport (STILT) model, *Journal of Geophysical Research*, 108(D16).
- Liu, J. J., et al. (2009), Analysis of the summertime buildup of tropospheric ozone abundances over the Middle East and North Africa as observed by the Tropospheric Emission Spectrometer instrument, *Journal of Geophysical Research*, 114(D05304).
- Liu, X.-H., et al. (2010), Ozone profile retrievals from the Ozone Monitoring Instrument, *Atmospheric Chemistry and Physics*, *10*: 2521-2537.
- Liu, Z., et al. (2008), CALIPSO lidar observations of the optical properties of Saharan dust: A case study of long-range transport, *Journal of Geophysical Research*, 113(D7): D07207.
- Logan, J. A., et al. (1999), Trends in the vertical distribution of ozone: A comparison of two analyses of Ozone Sonde data, *Journal of Geophysical Research*, *104*(D21): 26373-26399.
- Lu, Z., et al. (2010), Sulfur dioxide emissions in China and sulfur trends in East Asia since 2000, *Atmospheric Chemistry and Physics Discussions*, 10: 8657-8715.
- Martin, R. V., et al. (2006), Evaluation of space-based constraints on global nitrogen oxide emissions with regional aircraft measurements over and downwind of eastern North America, *Journal of Geophysical Research*, *111*(D15308).
- Martin, R. V. (2008), Satellite remote sensing of surface air quality, *Atmospheric Environment*, 42(34): 7823-7843.
- Matthias, V., et al. (2004), The vertical aerosol distribution over Europe: Statistical analysis of Raman lidar data from 10 EARLINET stations, *Journal of Geophysical Research*, *109*(D18201).
- Mauzerall, D. L., et al. (1998), Photochemistry in biomass burning plumes and implications for tropospheric ozone over the tropical South Atlantic, *Journal of Geophysical Research*, *103*(D7): 8401-8423.
- McConnell, J., et al. (2007), 20th-century industrial black carbon emissions altered Arctic climate forcing, *Science*, *317*: 1381-1384.

- McConnell, J. R., et al. (2002), A 250-year high-resolution record of Pb flux and crustal enrichment in central Greenland, *Geophysical Research Letters*, 29(23): 2130-2133.
- Mercier, G. (2000), Lead isotope composition and elemental abundances in atmospheric aerosols collected at Alert station (Canadien Arctic) in 1994-1995: Sources and trajectories, 55 pp, Universite du Quebec a Montreal.
- Methven, J., and B. Hoskins (1999), The Advection of High-Resolution Tracers by Low-Resolution Winds, Journal of the Atmospheric Sciences, 56(18): 3262-3285.
- Methven, J., et al. (2006), Establishing Lagrangian connections between observations within air masses crossing the Atlantic during the International Consortium for Atmospheric Research on Transport and Transformation, *Journal of Geophysical Research*, *111*(D23S62).
- Mickley, L. J., et al. (2001), Uncertainty in preindustrial abundance of tropospheric ozone: Implications for radiative forcing calculations, *Journal of Geophysical Research*, *106*(D4): 3389-3399.
- Mishchenko, M. I., et al. (2007), Long-Term Satellite Record Reveals Likely Recent Aerosol Trend, *Science*, 315(5818): 1543-.
- Moore, J., et al. (2006), Sulfate source inventories from a Svalbard ice core record spanning the Industrial Revolution, *Journal of Geophysical Research*, 111(D15307).
- Mukai, H., et al. (2001), Regional characteristics of sulfur and lead isotope ratios in the atmosphere at several Chinese urban sites, *Environmental Science & Technology*, *35*: 1064-1071.
- Norman, A. L., et al. (1999), Sources of aerosol sulphate at Alert: Apportionment using stable isotopes, *Journal* of *Geophysical Research*, 104: 11619-11631.
- Novak, M., et al. (2001), Temporal trends in the isotope signature of air-borne sulfur in Central Europe, *Environmental Science & Technology*, 35: 255-260.
- Nowak, J., et al. (2004), Gas-phase chemical characteristics of Asian emission plumes observed during ITCT 2K2 over the eastern North Pacific Ocean, *Journal of Geophysical Research*, *109*(D23S19).
- NRC (2010), Global sources of local pollution: an Assessment of long-range transport of key air pollutants to and from the United States, 234 pp, National Research Council, The National Academies, Washington, DC.
- Nriagu, J. O., et al. (1991), Origin of sulfur in Canadian arctic haze from isotope measurements, *Nature*, 349: 142-145.
- Oltmans, S. J., et al. (2006), Long-term changes in tropospheric ozone, *Atmospheric Environment*, 40(17): 3156-3173.
- Oltmans, S. J., et al. (2008), Background ozone levels of air entering the west coast of the US and assessment of longer-term changes, *Atmospheric Environment*, *42*(24): 6020-6038.
- Ordóñez, C. (2006), Trend analysis of ozone and evaluation of nitrogen dioxide satellite data in the troposphere over Europe, Swiss Federal Institute of Technology, Zurich.
- Osterberg, E., et al. (2008), Ice core record of rising lead pollution in the North Pacific atmosphere, *Geophysical Research Letters*, 35(L05810).
- Osthoff, H. D., et al. (2008), High levels of nitryl chloride in the polluted subtropical marine boundary layer, *Nature Geoscience*, *1*(5): 324-328.
- Owen, R. C., et al. (2006), An analysis of the mechanisms of North American pollutant transport to the Central North Atlantic lower free troposphere, *Journal of Geophysical Research*, 111: D23S58.
- Park, R. J., et al. (2004), Natural and transboundary pollution influences on sulfate-nitrate ammonium aerosols in the United States: Implications for policy, *Journal of Geophysical Research*, *109*(D15204).
- Parrish, D., et al. (2009), Increasing ozone in marine boundary layer inflow at the west coasts of North America and Europe, *Atmospheric Chemistry and Physics*, 9: 1303-1323.
- Parrish, D. D., et al. (1992), Indication of photochemical histories of Pacific air masses from measurements of atmospheric trace species at Point Arena, California, *Geophysical Research Letters*, 97(D14): 15883-15901.
- Parrish, D. D., et al. (1993), Export of North America ozone pollution to the North Atlantic Ocean, *Science*, 259(5100): 1436-1439.
- Parrish, D. D., et al. (2007), Effects of mixing on evolution of hydrocarbon ratios in the troposphere, *Journal of Geophysical Research*, *112*(D10S34).
- Parrish, D. D., et al. (2010), Impact of transported background ozone inflow on summertime air quality in a California ozone exceedance area, *Atmospheric Chemistry and Physics*, *10*: 10093-10109.
- Patris, N., et al. (2000), Isotopic composition of sulfur in size-resolved marine aerosols above the Atlantic Ocean, *Journal of Geophysical Research*, 105: 14449-14457.
- Patris, N., et al. (2002), First sulfur isotope measurements in central Greenland ice cores along the preindustrial and industrial periods, *Journal of Geophysical Research*, 107(D11): 4115-4125.
- Pfister, G. G., et al. (2006), Ozone production from the 2004 North American boreal fires, *Journal of Geophysical Research*, 111(D24S07).
- Pisso, I., et al. (2009), Estimation of mixing in the troposphere from Lagrangian trace gas reconstructions during long-range pollution plume transport, *Journal of Geophysical Research*, *114*(D1): 9301-9311.
- Pochanart, P., et al. (2003), Regional background ozone and carbon monoxide variations in remote Siberia/East Asia, *Journal of Geophysical Research*, *108*(D1): 4028-4045.
- Prather, M., et al. (2001), Atmospheric chemistry and greenhouse gases, in *Climate change 2001: The scientific basis: Contribution of Working Group 1 to the Third Assessment Report of the Intergovernmental Panel on Climate Change*, edited by J. T. Houghton, et al., 49 pp., Cambridge University Press, New York.
- Preunkert, S., et al. (2001), Sulfate trends in a Col du Dome (French Alps) ice core: A record of anthropogenic sulfate levels in the European midtroposphere over the twentieth century, *Journal of Geophysical Research*, *106*: 31991-32004.
- Preunkert, S., et al. (2003), A seasonally resolved alpine ice core record of nitrate: Comparison with anthropogenic inventories and estimation of preindustrial emissions of NO in Europe, *Journal of Geophysical Research*, *108*(D21): 4681-4690.
- Price, H. U., et al. (2004), Photochemistry, ozone production, and dilution during long-range transport episodes from Eurasia to the northwest United States, *Journal of Geophysical Research*, 109(D23S13).
- Prospero, J. M. (1979), Mineral and sea-salt aerosol concentrations in various ocean regions, *Journal of Geophysical Research*, 84(C2): 725-731.
- Prospero, J. M. (2001), The Atmosphere-Ocean Chemistry Experiment (AEROCE): Background and major accomplishments, *IGACtivities Newsletter*, 24.
- Prospero, J. M., et al. (2003), Long-term record of NSS-sulfate and nitrate in aerosols on Midway Island, 1981-2000: Evidence of increased (now decreasing?) anthropogenic emissions from Asia, *Journal of Geophysical Research*, 108(D14019).
- Putaud, J.-P., and F. Raes (2004), A European aerosol phenomenology 2: Chemical characteristics of particulate matter at kerbside, urban, rural and background sites in Europe, *Atmospheric Environment*, 38: 2579-2595.
- Quinn, P. K., et al. (2002), A 3-year record of simultaneously measured aerosol chemical and optical properties at Barrow, Alaska, *Journal of Geophysical Research*, *107*(D11): 4130-4145.
- Quinn, P. K., et al. (2007), Arctic haze: Current trends and knowledge gaps, Tellus, 59B: 99-114.
- Quinn, P. K., et al. (2009), Decadal trends in aerosol chemical composition at Barrow, Alaska: 1976-2008, *Atmospheric Chemistry and Physics*, 9(22): 8883-8888.

- Raes, F., et al. (2000), The 2nd Aerosol Characterization Experiment (ACE-2): General overview and main results, *Tellus Series B-Chemical and Physical Meteorology*, *52*(2): 111-125.
- Rahn, K. A., and R. J. McCaffrey (1980), On the origin and transport of the winter Arctic Aerosol, *Annals of the New York Academy of Sciences*, 338(1): 486-503.
- Rastigejev, Y., et al. (2010), Resolving intercontinental pollution plumes in global models of atmospheric transport, *Journal of Geophysical Research*, 115(D02302).
- Ravetta, F., et al. (2007), Long-range transport and tropospheric ozone variability in the western Mediterranean region during the Intercontinental Transport of Ozone and Precursors (ITOP-2004) campaign, *Journal of Geophysical Research*, *112*(D10S46).
- Real, E., et al. (2007), Processes influencing ozone levels in Alaskan forest fire plumes during long-range transport over the North Atlantic, *Journal of Geophysical Research*, *112*: D10S41.
- Real, E., et al. (2008), Lagrangian analysis of low altitude anthropogenic plume processing across the North Atlantic, *Atmospheric Chemistry and Physics*, 8: 7737-7754.
- Real, E., et al. (2010), Toward a novel high-resolution modeling approach for the study of chemical evolution of pollutant plumes during long-range transport, *Journal of Geophysical Research 115*(D12): D12302.
- Reidmiller, D. R., et al. (2009), Interannual variability of long-range transport as seen at the Mt. Bachelor observatory, *Atmospheric Chemistry and Physics*, 9(2): 557-572.
- Sauvage, B., et al. (2007), Quantification of the factors controlling tropical tropospheric ozone and the South Atlantic maximum, *Journal of Geophysical Research*, *112*(D09309).
- Savoie, D. L., et al. (2002), Marine biogenic and anthropogenic contributions to non-sea-salt sulfate in the marine boundary layer over the North Atlantic Ocean, *Journal of Geophysical Research*, 107(D18): 4356-4376.
- Schnadt Poberaj, C., et al. (2009), Long-term changes in UT/LS ozone between the late 1970s and the 1990s deduced from the GASP and MOZAIC aircraft programs and from ozonesondes, *Atmospheric Chemistry and Physics*, 9: 5343-5369.
- Schoeberl, M. R., et al. (2007), A trajectory-based estimate of the tropospheric ozone column using the residual method, *Journal of Geophysical Research*, *112*(D24S49).
- Schwarz, J. P., et al. (2006), Single-particle measurements of midaltitude black carbon and lightscattering aerosols from the boundary layer to the lower stratosphere, *Journal of Geophysical Research*, *111*(D16207).
- Schwarz, J. P., et al. (2008), Coatings and their enhancement of black carbon light absorption in the tropical atmosphere, *Journal of Geophysical Research*, *113*(D03203).
- Sharma, S., et al. (2006), Variations and sources of the equivalent black carbon in the high Arctic revealed by long-term observations at Alert and Barrow: 1989-2003, *Journal of Geophysical Research*, *111*(D14208).
- Shaw, G. E. (1975), The vertical distribution of atmospheric aerosols at Barrow, Alaska, Tellus, 27: 39-49.
- Shaw, G. E. (1995), The Arctic haze phenomenon, *Bulletin of the American Meteorological Society*, 76: 2403-2413.
- Sherrell, R. M., et al. (2000), Temporal variability of Cd, Pb, and Pb isotope deposition in central Greenland snow, *Geochemistry Geophysics Geosystems*, *1*(5): 1002-1023.
- Shotyk, W., et al. (2005), Predominance of industrial Pb in recent snow (1994-2004) and ice (1842-1996) from Devon Island, Arctic Canada, *Geophysical Research Letters*, *32*(L21814).
- Simmonds, P. G., et al. (2004), Significant growth in surface ozone at Mace Head, Ireland, 1987-2003, *Atmospheric Environment*, 38(28): 4769-4778.
- Simonetti, A., et al. (2004), Pb isotopic investigation of aircraft-sampled emissions from the Horne smelter (Rouyn, Quebec): Implications for atmospheric pollution in northeastern North America, *Geochimica et Cosmochimica Acta*, 68(16): 3285-3294.

- Singh, H. B., et al. (2006), Overview of the summer 2004 Intercontinental Chemical Transport Experiment -North America (INTEX-A), *Journal of Geophysical Research*, *111*(D24S01).
- Singh, H. B., et al. (2009), Chemistry and transport of pollution over the Gulf of Mexico and the Pacific: Spring 2006 INTEX-B campaign overview and first results, *Atmospheric Chemistry and Physics*, 9: 2301-2318.
- Sirois, A., and L. A. Barrie (1999), Arctic lower tropospheric aerosol trends and composition at Alert, Canada: 1980-1995, *Journal of Geophysical Research*, 104(D9): 11599-11618.
- Smit, H. G. J., et al. (2007), Assessment of the performance of ECC-ozonesondes under quasi-flight conditions in the environmental simulation chamber: Insights from the Juelich Ozone Sonde Intercomparison Experiment (JOSIE), Journal of Geophysical Research, 112(D19306).
- Staehelin, J., et al. (1994), Surface ozone trends at Arosa (Switzerland), Atmospheric Environment, 28: 75-87.
- Stevenson, D. S., et al. (2006), Multimodel ensemble simulations of present-day and near-future tropospheric ozone, *Journal of Geophysical Research*, 111(D08301).
- Stohl, A., and T. Trickl (1999), A textbook example of long-range transport: Simultaneous observations of ozone maxima of stratospheric and North America origin in the free troposphere over Europe, *Journal* of Geophysical Research, 104(D23): 30445-30462.
- Stohl, A., et al. (2002), On the pathways and timescales of intercontinental air pollution transport, *Journal of Geophysical Research*, 107(D23): 4684-4700.
- Stohl, A., et al. (2003), A backward modeling study of intercontinental pollution transport using aircraft measurements, *Journal of Geophysical Research*, 108(D12): 4370-4387.
- Stohl, A., et al. (2005), Technical note: The Lagrangian particle dispersion model FLEXPART version 6.2, *Atmospheric Chemistry and Physics*, 5: 2461-2474.
- Stohl, A., et al. (2007a), Arctic smoke record high air pollution levels in the European Arctic due to agricultural fires in Eastern Europe in spring 2006, *Atmospheric Chemistry and Physics*, 7: 511-534.
- Stohl, A., et al. (2007b), Aircraft measurements over Europe of an air pollution plume from Southeast Asia aerosol and chemical characterization, *Atmospheric Chemistry and Physics*, 7(3): 913-937.
- Streets, D. G., et al. (2000), Sulfur dioxide emissions in Asia in the period 1985 -1997, *Atmospheric Environment*, *34*(26): 4413-4424.
- Sturges, W. T., et al. (1993), Stable lead isotope ratios in Alaskan arctic aerosols, *Atmospheric Environment*, 27(17-18): 2865-2871.
- Takami, A., et al. (2006), Transport of anthropogenic and dust aerosol observed at Cape Hedo, Okinara, *Journal* of Aerosol Research, 21(4): 341-347.
- Tanimoto, H. (2009), Increase in springtime tropospheric ozone at a mountainous site in Japan for the period 1998-2006, *Atmospheric Environment*, 43: 1358-1363.
- Tanimoto, H., et al. (2009), Asian anthropogenic emissions and decadal trends in springtime tropospheric ozone over Japan: 1998-2007, *Geophysical Research Letters*, *36*(L23802).
- Thomas, G. E., et al. (2010), Validation of the GRAPE single view aerosol retrieval for ATSR-2 and insights into the long term global AOD trend over the ocean, *Atmospheric Chemistry and Physics*, *10*(10): 4849-4866.
- Thouret, V., et al. (2006), Tropopause referenced ozone climatology and inter-annual variability (1994-2003) from the MOZAIC programme, *Atmospheric Chemistry and Physics*, *6*(4): 1033-1051.
- Torres, O., et al. (2002), A long-term record of aerosol optical depth from TOMS observations and comparison to AERONET measurements, *Journal of Atmospheric Science*, *59*(3): 398-413.
- Torres, O., et al. (2007), Aerosols and surface UV products from Ozone Monitoring Instrument observations: An overview, *Journal of Geophysical Research*, *112*(D24S47).

- Trickl, T., et al. (2003), Intercontinental transport and its influence on the ozone concentrations over central Europe: Three case studies, *Journal of Geophysical Research*, *108*(D12): 8530-8552.
- Turnbull, J. C., et al. (2009), Spatial distribution of 14CO2 across Eurasia: Measurements from the TROICA-8 expedition, *Atmospheric Chemistry and Physics*, *9*: 175-187.
- Val Martin, M., et al. (2006), Significant enhancements of nitrogen oxides, black carbon, and ozone in the North Atlantic lower free troposphere resulting from North American boreal wildfires, *Journal of Geophysical Research*, 111(D23S60).
- Val Martin, M., et al. (2008), Large-scale impacts of anthropogenic pollution and boreal wildfires on the nitrogen oxides over the central North Atlantic region, *Journal of Geophysical Research*, *113*(D17308).
- Van Dingenen, R., and F. Raes (2004), A European aerosol phenomenology 1: Physical characteristics of particulate matter at kerbside, urban, rural and background sites in Europe, *Atmospheric Environment*, 38: 2561-2577.
- van Donkelaar, A., et al. (2008), Analysis of aircraft and satellite measurements from the Intercontinental Chemical Transport Experiment (INTEX-B) to quantify long-range transport of East Asian sulfur to Canada, *Atmospheric Chemistry and Physics*, 8: 2999-3014.
- VanderWal, R., et al. (2010), Fingerprinting soot (towards source identification): Physical structure and chemical composition, *Aerosol Science*, *41*: 108-117.
- Vestreng, V. (2003), Review and revision. Emission data reported to CLRTAP, 134 pp, Norwegian Meteorological Institute, Oslo. MSC-W Technical Report 1/03. http://emep.int/publ/reports/2003/mscw_note_1_2003.pdf
- Veysseyre, A. M., et al. (2001), Tracing the origin of pollution in French Alpine snow and aerosols using lead isotopic ratios, *Environmental Science & Technology*, *35*(22): 4463-4469.
- Viana, M. T., et al. (2008), Source apportionment of particulate matter in Europe: A review of methods and results, *Journal of Aerosol Science*, 39: 827-849.
- Volz, A., and D. Kley (1988), Evaluation of the Montsouris series of ozone measurements made in the 19th century, *Nature*, 332(6161): 240-242.
- Vukicevic, T., and P. Hess (2000), Analysis of tropospheric transport in the Pacific Basin using the adjoint technique, *Journal of Geophysical Research*, 105(D6): 7213-7230.
- Wadleigh, M. A., et al. (2001), Areal distribution of sulphur and oxygen isotopes in sulphate of rain over eastern North America, *Journal of Geophysical Research*, *106*(D18): 20883-20895.
- Wadleigh, M. A. (2004), Sulphur isotopic composition of aerosols over the western North Atlantic Ocean, Canadian Journal of Fisheries and Aquatic Sciences, 61(5): 817-825.
- Wandinger, U., et al. (2004), Air mass modification over Europe: EARLINET aerosol observations from Wales to Belarus, *Journal of Geophysical Research*, 109(D24205).
- Wang, T., et al. (2009), Increasing surface ozone concentrations in the background atmosphere of Southern China, 1994-2007, *Atmospheric Chemistry and Physics*, 9: 6217-6227.
- Warneke, C., et al. (2007), Determination of urban volatile organic compound emission ratios and comparison with an emissions database, *Journal of Geophysical Research*, *112*(D10S47).
- Weiss-Penzias, P., et al. (2007), Quantifying Asian and biomass burning sources of mercury using the Hg/CO ratio in pollution plumes observed at the Mount Bachelor observatory, *Atmospheric Environment*, 41(21): 4366-4379.
- Wespes, C., et al. (2009), Global distributions of nitric acid from IASI/MetOP measurements, *Atmospheric Chemistry and Physics*, 9: 7949-7962.
- WHO (2006), Air quality guidelines: Global update 2005: Particulate matter, ozone, nitrogen dioxide, and sulfur dioxide, 22 pp, World Health Organization, Geneva. WHO/SDE/PHE/OEH/06.02. http://whqlibdoc.who.int/hq/2006/WHO SDE PHE OEH 06.02 eng.pdf

Wilkening, K. E., et al. (2000), Trans-Pacific air pollution, Science, 290: 65-67.

- Wittrock, F., et al. (2006), Simultaneous global observations of glyoxal and formaldehyde from space, *Geophysical Research Letters*, 33(L16804).
- Worden, H. M., et al. (2007), Comparison of Tropospheric Emission Spectrometer (TES) ozone profiles to ozonesondes: Methods and initial results, *Journal of Geophysical Research*, 112(D03309).
- Worden, J. R., et al. (2007), Improved tropospheric ozone profile retrievals using OMI and TES radiances, *Geophysical Research Letters*, 34(L01809).
- Yalcin, K., and C. Wake (2001), Anthropogenic signals recorded in an ice core from Eclipse Icefield, Yukon Territory, Canada, *Geophysical Research Letters*, 28: 4487-4490.
- Yang, J., et al. (2009), Isotopic evidences for provenance of East Asian dust, *Atmospheric Environment*, 43: 4481-4490.
- Yang, M., et al. (2006), Microparticle content records of the Dunde ice core and dust storms in northwestern China, *Journal of Asian Earth Sciences*, 27: 223-229.
- Yasunari, T. J., et al. (2007), Intra-annual variations in atmospheric dust and tritium in the North Pacific region detected from an ice core from Mount Wrangell, Alaska, *Journal of Geophysical Research*, *112*(D10208).
- Yasunari, T. J., and K. Yamazaki (2009), Impacts of Asian dust storm associated with the stratosphere-totroposphere transport in the spring of 2001 and 2002 on dust and tritium variations in Mount Wrangell ice core, Alaska, *Atmospheric Environment*, *43*: 2582-2590.
- Yu, H., et al. (2008), A satellite-based assessment of transpacific transport of pollution aerosol, *Journal of Geophysical Research*, 113(D14S12).
- Zbinden, R. B., et al. (2006), Mid-latitude tropospheric ozone columns from the MOZAIC program: Climatology and interannual variability, *Atmospheric Chemistry and Physics*, 6: 863-881.
- Zdanowicz, C., et al. (2006), Asian dustfall in the St. Elias Mountains, Yukon, Canada, *Geochimica et Cosmochimica Acta*, 70: 3493-3507.
- Zhang, L., et al. (2008), Transpacific transport of ozone pollution and the effect of recent Asian emission increases on air quality in North America: An integrated analysis using satellite, aircraft, ozonesonde, and surface observations, *Atmospheric Chemistry and Physics*, 8(20): 6117-6136.
- Zhang, L., et al. (2009), Intercontinental source attribution of ozone pollution at western U.S. sites using an adjoint method, *Geophysical Research Letters*, *36*(L11810).
- Zheng, S., et al. (2009), Mineralogical characteristics of soil dust from source regions in northern China, *Particuology*, 7: 507-512.
- Ziemke, J. R., et al. (2006), Tropospheric ozone determined from Aura OMI and MLS: Evaluation of measurements and comparison with the global modeling initiative's chemical transport model, *Journal* of Geophysical Research, 111(D19303).

Appendix A

Editors, Authors, & Reviewers

Contributor	Organization / Affiliation	Country
Elizabeth Ainsworth	Agricultural Research Service, USDA	USA
Hajime Akimoto	Asia Center for Air Pollution Research	Japan
Susan C. Anenberg	University of North Carolina, Chapel Hill &	USA
	Environmental Protection Agency	
Ahmareen Arif	Air University	Pakistan
Steve Arnold	University of Leeds	United Kingdom
Mike Ashmore	Stockholm Environment Institute, York	United Kingdom
Richard Atkinson	St. George's University of London	United Kingdom
Marianne Bailey	Environmental Protection Agency	USA
Paul Bartlett	St. Peter's College & CUNY	USA
William Battye	EC/R Inc.	USA
Nicolas Bellouin	Met Office Hadley Centre	United Kingdom
Terry Bidleman	Environment Canada	Canada
Knut Breivik	Norwegian Institute for Air Research	Norway
O. Russell Bullock	Environmental Protection Agency	USA
Greg Carmichael	University of Iowa	USA
Elton Chan	Environment Canada	Canada
Gao Chen	National Aeronautics & Space Administration	USA
Mian Chin	National Aeronautics & Space Administration	USA
Sergio Cinnirella	CNR Institute of Atmospheric Pollution	Italy
	Research	
Aaron Cohen	Health Effects Institute	USA
William Collins	Met Office Hadley Centre, Exeter	United Kingdom
Owen Cooper	National Oceanographic & Atmospheric	USA
	Administration	
Elizabeth Corbitt	Harvard University	USA
Daniel Cossa	IFremer Centre de Mediterranee	France
Ashu Dastoor	Environment Canada	Canada
John Dawson	Environmental Protection Agency	USA
Pierre Delmelle	University of York	United Kingdom
Hugo Denier van der Gon	I NO Built Environment and Geosciences	Netherlands
Frank Dentener	Joint Research Centre – European Commission	European Community
Richard Derwent	rdscientific, Newbury	United Kingdom
Ruth Doherty	University of Edinburgh	United Kingdom
Pat Dolwick	Environmental Protection Agency	USA
Aurelien Dommergue	Universite Joseph Fourier-Grenoble	France
Robert A. Duce	Texas A&M University	USA
Sergey Dutchak	EMEP/MSC-E	Russia
Kristie L. Ebi	IPCC WGII-TSU, Carnegie Institution for	USA
	Science	0
Ralf Ebinghaus	Helmholtz-Zentrum Geesthacht - Institute for	Germany
D 101 1	Coastal Research	
David Edwards	National Center for Atmospheric Research	USA
Lisa Emberson	Stockholm Environment Institute, York	United Kingdom
David Evers	Biodiversity Research Institute	USA
Nasreen Farah	Hydrocarbon Development Institute of Pakistan	Pakıstan
Xinbin Feng	Chinese Academy of Science	China

Contributor	Organization / Affiliation	Country
Arlene Fiore	National Oceanographic & Atmospheric	USA
	Administration	
Gerd Folberth	Met Office Hadley Centre	United Kingdom
Hans Friedli	National Center for Atmospheric Research	USA
Joshua Fu	University of Tennessee	USA
Jürg Fuhrer	Agroscope Research Station ART	Switzerland
Savitri Garivait	JGSEE - King Mongkut's University of	Thailand
	Technology Thonburi	
Sunling Gong	Environment Canada	Canada
Claire Granier	Service d'Aéronomie, Centre National de la	France
	Recherche Scientifique (CNRS)	
Doug Grano	EC/R Inc.	USA
Ramon Guardans	Ministry of the Environment and Rural and	Spain
	Marine Affairs	*
Alex Guenther	National Center for Atmospheric Research	USA
Alexey Gusev	EMEP/MSC-E	Russia
Mae Gustin	University of Nevada, Reno	USA
Kimberly Hageman	University of Otago	New Zealand
Simon Hales	University of Otago, Wellington	New Zealand
Crispin Halsall	Lancaster University	United Kingdom
Tom Harner	Environment Canada	Canada
Ian M.Hedgecock	CNR Institute of Atmospheric Pollution	Italy
C C	Research	-
Peter Hess	Cornell University	USA
Kevin Hicks	Stockholm Environment Institute &	United Kingdom
	University of York	C
Anne Hollander	Radboud University Nijmegen	Netherlands
Tracey Holloway	University of Wisconsin - Madison	USA
Christopher Holmes	Harvard University	USA
Ivan Holoubek	Masaryk University	Czech Republic
Hayley Hung	Environment Canada	Canada
Ilia Ilyin	EMEP/MSC-E	Russia
Lyatt Jaeglé	University of Washington	USA
Dan Jaffe	University of Washington - Bothell	USA
Liisa Jantunen	Environment Canada	Canada
S. Gerard Jennings	National University of Ireland, Galway	Ireland
Jan Eiof Jonson	Norwegian Meteorological Institute	Norway
Gerlinde Jung	University of Bremen	Germany
Roland Kallenborn	Norwegian Institute for Air Research	Norway
Maria Kanakidou	University of Crete	Greece
Terry Keating	Environmental Protection Agency	USA
Gerald J. Keeler	University of Michigan	USA
Zbigniew Klimont	International Institute for Applied Systems	Austria
-	Analysis	
Kazuhiko Kobayashi	The University of Tokyo	Japan
Dorothy Koch	Department of Energy	USA
Hans Herbert Kock	Helmholtz-Zentrum Geesthacht - Institute for	Germany
	Coastal Research	-
Charles E. Kolb	Aerodyne Research, Inc.	USA
David Krabbenhoft	United States Geological Survey	USA
Paolo Laj	Laboratoire de Glaciologie, Centre National de	France
-	la Recherche Scientifique (CNRS)	

Contributor	Organization / Affiliation	Country
Yun-Fat Lam	University of Tennessee	USA
Jean-Francois Lamarque	National Center for Atmospheric Research	USA
Gerhard Lammel	Max Planck Institute for Chemistry	Germany
Kathy Law	Service d'Aéronomie, CNRS	France
Leonard Levin	Electric Power Research Institute	USA
Yi-Fan Li	Environment Canada	Canada
Che-Jen Lin	Lamar University	USA
Meiyun Lin	Princeton University & National	USA
	Oceanographic & Atmospheric Administration	
Junfeng Liu	Princeton University & National	USA
-	Oceanographic & Atmospheric Administration	
Zifeng Lu	Argonne National Laboratory	USA
Jianmin Ma	Environment Canada	Canada
Robie Macdonald	Fisheries and Oceans Canada	Canada
Matthew MacLeod	Swiss Federal Institute of Technology	Switzerland
Greet Maenhout	Joint Research Centre - European Commission	European Community
Randall Martin	Dalhousie University	Canada
Robert Mason	University of Connecticut	USA
Denise Mauzerall	Princeton University	USA
David McCabe	Clean Air Task Force	USA
Lina Mercado	Centre for Ecology & Hydrology	United Kingdom
John Methven	University of Reading	United Kingdom
Torsten Meyer	University of Toronto	Canada
Gina Mills	Centre for Ecology & Hydrology	United Kingdom
Manju Mohan	Indian Institute of Technology (IIT), Delhi	India
Paul Monks	University of Leicester	United Kingdom
Arun B. Mukherjee	University of Helsinki	Finland
Toshimasa Ohara	National Institute for Environmental Studies	Japan
Koyo Ogasawara	IDEA Consultants, Inc	Japan
Elisabeth G.Pacyna	Norwegian Institute for Air Research	Norway
Jozef Pacyna	Norwegian Institute for Air Research	Norway
Li Pan	Lamar University	USA
Damian Panasiuk	Norwegian Institute for Air Research (NILU- Polska)	Poland
David Parrish	National Oceanographic & Atmospheric Administration	USA
Stuart Penkett	University of East Anglia	United Kingdom
Nicola Pirrone	CNR Institute of Atmospheric Pollution	Italy
	Research	2
Håkan Pleijel	University of Gothenburg	Sweden
Pruek Pongprueksa	Lamar University	USA
Joe Prospero	University of Miami	USA
Patricia Quinn	National Oceanographic & Atmospheric	USA
	Administration	
David Reidmiller	University of Washington - Bothell	USA
Lorraine Remer	National Aeronautics & Space Administration	USA
Glenn Rice	Harvard University	USA
Sergiu Robu	Academy of Sciences of Moldova	Moldova
Andrew Ryzhkov	Environment Canada	Canada
Michael Sanderson	Met Office Hadley Centre	United Kingdom
Rich Scheffe	Environmental Protection Agency	USA
David Schmeltz	Environmental Protection Agency	USA

Contributor	Organization / Affiliation	Country
Michael Schulz	Norwegian Meteorological Institute	Norway
Christian Seigneur	Centre d'Enseignement et de Recherche en	France
	Environnement Atmosphérique	
Noelle Eckley Selin	Massachusetts Institute of Technology	USA
Victor Shatalov	EMEP/MSC-E	Russia
Drew Shindell	National Aeronautics & Space Administration	USA
Staci Simonich	Oregon State University	USA
Stephen Sitch	University of Leeds	United Kingdom
Henrik Skov	National Environmental Research Institute	Denmark
Steven Smith	Pacific Northwest National Laboratory	USA
Francesca Sprovieri	CNR Institute of Atmospheric Pollution	Italy
	Research	
Johannes Staehelin	Institute for Atmospheric and Climate Science	Switzerland
David S. Stevenson	University of Edinburgh, School of Geosciences	United Kingdom
Andreas Stohl	Norwegian Institute for Air Research (NILU)	Norway
David Stone	Environment Canada (retired)	Canada
David Streets	Argonne National Laboratory	USA
Yushan Su	Environment Canada	Canada
Elsie Sunderland	Harvard University	USA
Kyrre Sundseth	Norwegian Institute for Air Research	Norway
Norivuki Suzuki	National Institute for Environmental Studies	Japan
Andy Sweetman	Lancaster University	United Kingdom
Shoaib-Raza Syed	University of Strasbourg	France
Akinori Takami	National Institute for Environmental Studies	Japan
Hiroshi Tanimoto	National Institute for Environmental Studies	Japan
Shu Tao	Peking University	China
Jochen Theloke	University of Stuttgart	Germany
Valerie Thouret	Laboratoire d'Aérologie, CNRS	France
Oleg Travnikov	EMEP/MSC-E	Russia
Thomas Trickl	Karlsruher Institut für Technologie	Germany
Juha-Pekka Tuovinen	Finnish Meteorological Institute	Finland
Solene Turquety	Laboratoire de Météorologie Dynamique, CNRS	France
Harry Vallack	University of York	United Kingdom
John van Aardenne	European Environment Agency	European Community
Rita van Dingenen	Joint Research Centre – European Commission	European Community
Jun Wang	University of Nebraska, Lincoln	USA
Yuxuan Wang	Tsinghua University	China
Peter Weiss	Umweltbundesamt	Austria
J. Jason West	University of North Carolina, Chapel Hill	USA
Jeffrey L. West	Environmental Protection Agency	USA
Oliver Wild	Lancaster University	United Kingdom
Wilfried Winiwarter	International Institute of Applied Systems	Austria
Hongbin Yu	University of Maryland & National	USA
	Aeronautics & Space Administration	C 1
Christian Zdanowicz	Natural Resources Canada	Canada
Yang Zhang	North Carolina State University	USA
Jerry Ziemke	National Aeronautics & Space Administration	USA
Andre Zuber	European Commission	European Community