Hemispheric Transport of Air Pollution 2010

Part D: Answers to Policy-Relevant Science Questions

Air Pollution Studies No. 20

UNITED NATIONS

ECONOMIC COMMISSION FOR EUROPE
Geneva

HEMISPHERIC TRANSPORT OF AIR POLLUTION 2010

PART D: ANSWERS TO POLICY-RELEVANT SCIENCE QUESTIONS

AIR POLLUTION STUDIES No. 20

Prepared by the Task Force on Hemispheric Transport of Air Pollution
acting within the framework of the
Convention on Long-range Transboundary Air Pollution

UNITED NATIONS
New York and Geneva, 2010

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Contents

Appendix

Tables

Figures

Chemical Symbols, Acronyms and Abbreviations

Chemical Abbreviations

C – carbon
CO – carbon monoxide
CO_2 – carbon dioxide
CH_4 – methane
DDT -- dichlorodiphenyltrichloroethane
HCH – hexachlorohexane
H_2SO_4 – sulphuric acid
HCB – hexachlorobenzene
HCHO – formaldehyde
Hg – mercury
Hg^0 – elemental mercury
Hg^{II} – oxidized, ionic mercury
Hg^P – mercury bound to suspended particulate matter
MeHg – methyl mercury
NH_3 – ammonia
NO_2 – nitrogen dioxide
NO_3 – nitrate
NO_x – nitrogen oxides
NO_y – total inorganic oxidized nitrogen
N_2O_5 -- dinitrogen pentoxide
Nr – total reactive nitrogen (including NO_y and NH_x)
O_3 – ozone
PAH – polycyclic aromatic hydrocarbon
PBDE – pentabromodiphenyl ether
PCB – polychlorinated biphenyl
PCDD/F – pentachlorodibenzodioxins and furans
PFOA – perfluorooctanoic acid
PFOS – perfluorooctane sulfonate
SO_2 – sulphur dioxide
SO_4 –sulphate
SO_x – anthropogenic sulphur oxides (combination of SO_2 and SO_4)
$nss-SO_4$ – non-seasalt sulphate

Acronyms and Abbreviations

ABL – Atmospheric Boundary Layer
AC&C – Atmospheric Chemistry and Climate (an initiative of the International Geosphere-Biosphere Program-World Climate Research Program)
AeroCom – Aerosol Comparisons between Observations and Models (a global aerosol model intercomparison project)
AIRS – Atmospheric Infrared Sounder
AMDEs – Atmospheric Mercury Depletion Events
AOT-40 – Accumulated Ozone exposure over a Threshold of 40 parts per billion
asl – above sea level
CALIPSO – Cloud-Aerosol Lidar and Infrared Pathfinder Satellite Observations
CASTNet – Clean Air Status and Trends Network
CLRTAP – Convention on Long Range Transboundary Air Pollution

EA – East Asia
EANET – Acid Deposition Monitoring Network in East Asia
EBAS-HTAP – observation database developed by NILU
EDGAR – Emissions Database for Global Atmospheric Research
EMEP – Cooperative Programme for Monitoring and Evaluation of the Long-range Transmission of Air Pollutants in Europe
EMEP-CCC – EMEP Chemical Coordinating Centre
EU -- Europe
GAW – Global Atmospheric Watch Programme (within WMO)
GEO – Group on Earth Observations
GEOSS – Global Earth Observing System of Systems
HTAP – Hemispheric Transport of Air Pollution
ICAO – International Civil Aviation Organization
IGAC – International Global Atmospheric Chemistry
IGACO – Integrated Global Atmospheric Chemistry Observations
IGBP – International Geosphere-Biosphere Programme (part of the International Council of Scientific Unions)
IMO – International Maritime Organization
IPCC – Intergovernmental Panel on Climate Change
K – degrees Kelvin
km – kilometres
LRTAP – Long-range Transboundary Air Pollution
NA – North America
NASA – National Aeronautics and Space Administration
ng m^{-3} – nanograms per cubic metre
NILU – Norweigan Institute for Air Research
PAN – Peroxyacetyl Nitrate
PJ – Polar Jet stream
PM – Particulate Matter
POM – Particulate Organic Matter
POPs – Persistent Organic Pollutants
ppbv – parts per billion by volume
RAIR – Relative Annual Intercontinental Response
RCP – Representative Concentration Pathways
S/R – source-receptor
SA – South Asia
SJ – Subtropical Jet stream
SRES – Special Report on Emissions Scenarios
SUM06 – the sum of all hourly ozone concentrations greater than 0.06 parts per million
TF HTAP – Task Force on Hemispheric Transport of Air Pollutants
TP – process tracer study
$\mu g/m^3$ – micrograms per cubic metre
UNECE – United Nations Economic Commission for Europe
UNEP – United Nations Environmental Programme
UNFCCC – United Nations Framework Convention on Climate Change
VOCs – Volatile Organic Compounds
WCB – Warm Conveyor Belt
WMO – World Meteorological Organization

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 Long-range 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 (O_3), 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), mercury (Part B), and persistent organic pollutants (Part C). This 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.

We would also like to thank the other task forces and centres under the LRTAP Convention as well as the staff of the Convention secretariat and EC/R Inc., who supported our work and the production of the report.

André Zuber and Terry Keating
Co-chairs of the Task Force on Hemispheric Transport of Air Pollution

Answers to Policy-Relevant Science Questions

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Introduction

Ozone (O_3), particulate matter (PM), mercury (Hg), and persistent organic pollutants (POPs) are significant environmental problems in many regions of the world. For each of these pollutants, observed concentrations or deposition at any given location can be thought of as composed of several different fractions, one of which is related to the intercontinental atmospheric transport of anthropogenic emissions into the air. Other fractions may be associated with natural emission sources or local and regional anthropogenic sources. Each fraction differs in terms of the emission sources that contribute to it, the temporal and spatial variability of the contribution, and the potential for and sensitivity to emission controls.

The Task Force on Hemispheric Transport of Air Pollution (TF HTAP) was organized under the Convention on Long-range Transboundary Air Pollution (LRTAP Convention) to improve the understanding of how the intercontinental transport of air pollutants across the Northern Hemisphere affects the achievement of air quality and other environmental goals. The TF HTAP started by articulating a series of policy-relevant science questions to guide its work and then organized a series of cooperative projects and assessment activities to address these questions. These guiding questions, which have evolved a little over the last five years, are as follows:

1. What is the observational evidence for the intercontinental transport of O_3, PM, Hg, and POPs in the Northern Hemisphere?

2. What are the main processes that drive these intercontinental flows and determine their magnitudes?

3. What do current models tell us about the contribution of intercontinental or global flows to concentrations and deposition in the different regions of the Northern Hemisphere and how changes in current emissions in one region affect air pollution concentrations or deposition in another region?

4. What is the contribution of these intercontinental or global flows to impacts on human health, natural and agricultural ecosystems, and near-term climate change?

5. How may the source-receptor relationships change over the next 20 to 40 years due to changes in emissions and climate change?

6. How well can we represent the processes that affect these intercontinental or global flows of air pollutants in quantitative models?

7. What efforts are needed to develop a system of observations, emissions, and models to better understand and track these flows?

8. What are the potential benefits of further international cooperation to address intercontinental transport of air pollution and how might this cooperation be structured?

In this volume (Part D of HTAP 2010), we provide summary answers to these questions, drawing upon the more detailed information presented in Parts A, B, and C of the report. The summary information presented here is provided without references to the scientific literature. For the supporting analysis that underlies the findings presented here, the reader is encouraged to refer to the technical discussions in Parts A, B, and C which contain extensive bibliographies.

The main messages that emerge from the answers to these questions are as follows:

- O_3, PM, Hg, and POPs are significant environmental problems in many regions of the world.

- Mitigation of intercontinental transport is not a substitute for emission reductions at the local and regional scale. In most cases, concentrations within a source region are more sensitive to emission changes within that region.

- However, without further international cooperation to mitigate intercontinental flows of air pollution, many nations are not able currently to meet their own goals and objectives for protecting public health and environmental quality.

- With changing global emissions, it is likely that over the next 20 to 40 years it will become even more difficult for nations to meet their own environmental policy objectives without international cooperation to address transboundary and intercontinental flows of air pollution.

- Cooperation to decrease emissions that contribute to intercontinental transport of air pollution has significant benefits for both source and receptor countries.

- There are formal intergovernmental forums for furthering global cooperation to mitigate sources of POPs and Hg. However, there is no existing international agreement addressing global or intercontinental flows of O_3 and PM pollution across the Northern Hemisphere.

1. **What is the observational evidence for the intercontinental transport of ozone, particulate matter, mercury, and persistent organic pollutants in the Northern Hemisphere?**

Observations from instruments and collectors on the ground, connected to balloons, in aircraft, or on satellites provide a wealth of evidence that concentrations and deposition of O_3, PM, Hg, and POPs are influenced by atmospheric transport between continents and, in some cases, around the globe.

Particulate Matter

Some of the most tangible evidence for intercontinental transport of air pollution comes from satellite images of PM concentrations, often associated with forest or grass fires and windblown soil dust storms, which travel across oceans and continents in visible plumes. Within the past few years satellites have begun to provide quantitative information on intercontinental transport of PM pollution associated with these events, as well as for more typical emissions from fossil fuel combustion. These data include estimates of the amount of pollution transported, the altitude of transport and in some cases, aerosol properties, such as their ability to reflect or absorb solar radiation. Figure 1.1 presents observations of trans-Atlantic transport of Saharan dust and open biomass burning smoke from CALIPSO (Cloud-Aerosol Lidar and Infrared Pathfinder Satellite Observations).
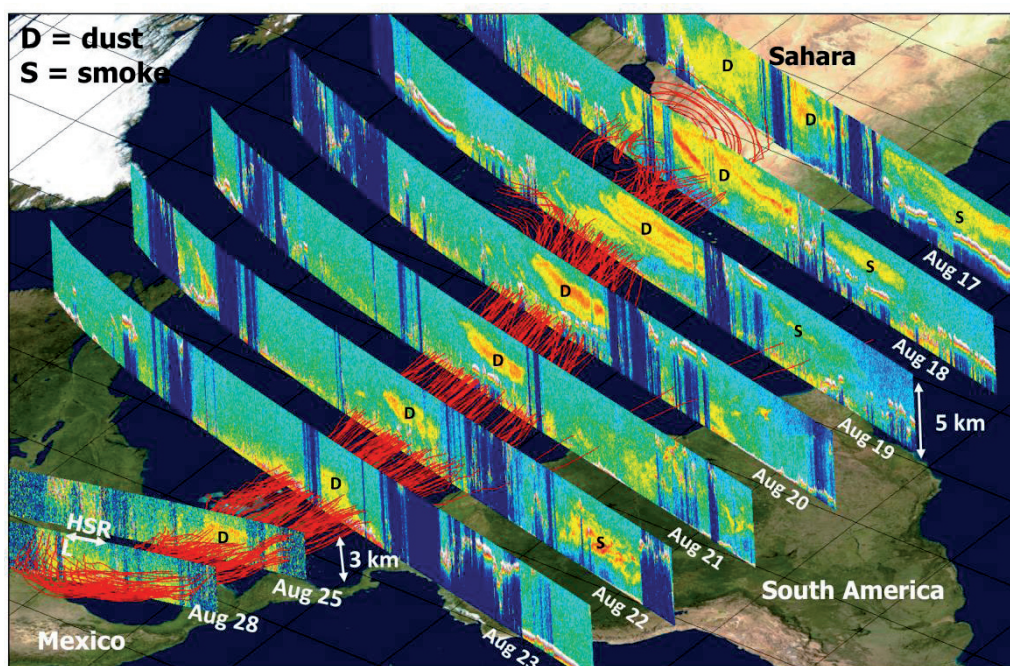
Figure 1.1. A dust event that originated in the Sahara desert on 17 August 2007 and was transported to the Gulf of Mexico. Red lines represent back trajectories indicating the transport track of the dust event. Vertical images are 532 nm attenuated backscatter coefficients measured by CALIPSO when passing 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.]

Ground-based lidar networks and mountain top measurement sites in Europe, North America and Asia provide large continuous data sets that characterize the frequency of occurrence of aerosol transport events, the meteorological conditions responsible for them, and important information on aerosol properties. Evidence of intercontinental transport is also provided in the form of long-term trends in surface concentration and wet deposition observations from remote islands and other remote locations, which in some cases are comparable to the emission trends in upwind areas. Some *in situ* measurements have also illuminated the importance of secondary aerosol formation from precursors

transported from other regions. Observations of the trace element composition and stable isotope ratios of PM have been useful in assessing the importance of natural and anthropogenic sources and studying processing of aerosols in transit.

Ozone

For O_3, evidence of intercontinental transport comes from direct O_3 measurements as well as measurements of precursor gases. Plumes of elevated O_3 have been observed in the free troposphere and at high elevation sites. Most importantly, an increasing trend in baseline O_3 concentrations, i.e., concentrations in air masses without the contribution from local anthropogenic emissions, has been measured consistently at a number of remote sites across the Northern Hemisphere, shown in Figure 1.2. Measurements suggest that during the latter half of the 20th century, concentrations of O_3 at northern mid-latitudes increased by a factor of two or more. It is likely that much of this change is due to increases in anthropogenic emissions of O_3 precursors.
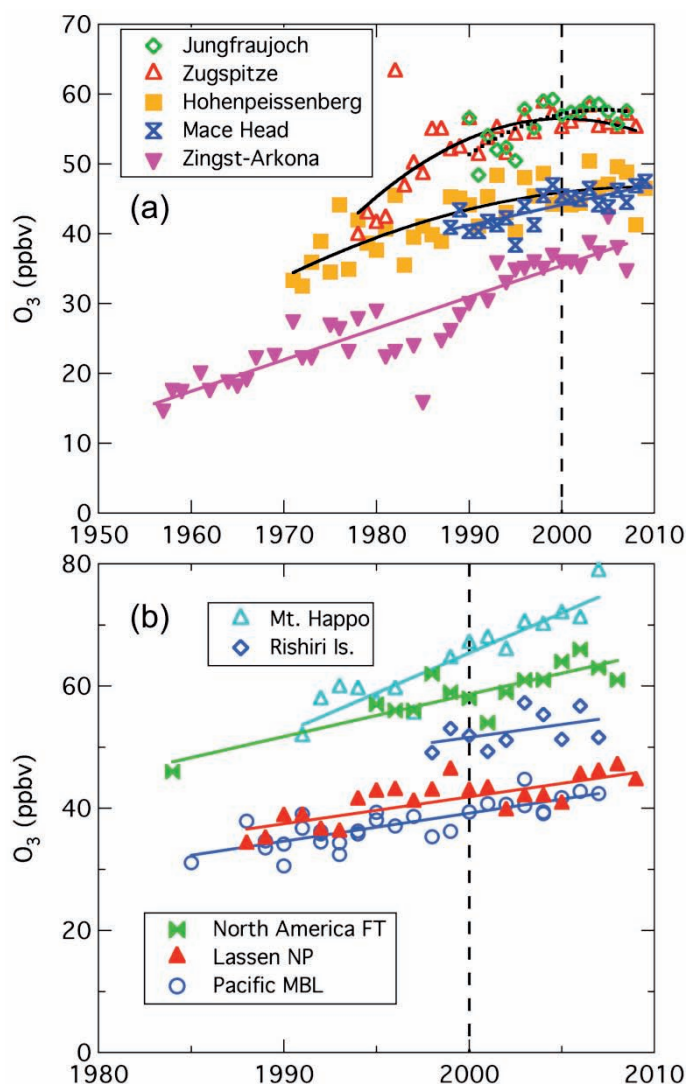
Figure 1.2. Springtime trends in O_3 concentrations measured in (a) Europe and (b) western North America and Japan. The lines (in colour) indicate the linear regressions to the data, and the curves (in black) indicate quadratic polynomial fits to the three central European sites over the time span of 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.

4

Within the limits of the measurement records, the increase has been comparable throughout all longitudes, and has occurred in all seasons. More recently, more rapid increases have been observed downwind of eastern Asia in the free troposphere, whereas the increases within the boundary layer of central Europe and North America have slowed down. Measurements at some locations on the western coasts of Europe and North America clearly show that trans-oceanic air flows can carry O_3 concentrations that approach or exceed air quality standards and objectives, and that air can mix to the surface and contribute substantially to air quality standard violations. This is particularly noticeable in areas with low emissions. The impact on surface air quality depends upon vertical mixing of air into the boundary layer, which is enhanced by complex topography.

Mercury

In the atmosphere, Hg is found in the form of elemental Hg (Hg^0), and ionic Hg in both gas and particle form. While PM is often removed from the atmosphere by wet or dry deposition within a matter of days to weeks and O_3 may last weeks to months, Hg^0 has an average residence time in the atmosphere of six months to a year. This is sufficient time to become relatively well-mixed, creating Hg^0 baseline concentrations of 1.5 to 1.7 nanograms per cubic metre (ng m^{-3}) in the Northern Hemisphere and 1.1 to 1.3 ng m^{-3} in the Southern Hemisphere (at sea level). This inter-hemispheric gradient has been observed consistently during ocean cruises.

Hg^0 is removed from the atmosphere after oxidation to ionic Hg, which may exist as a gas or incorporated into aerosols. Whether as a gas or an aerosol, ionic Hg deposits relatively quickly and is estimated to comprise less than five percent of the total atmospheric loading of Hg. Thus, the potential for intercontinental transport of Hg emissions depends on the chemical form in which it is emitted. Natural emissions are almost entirely Hg^0; however, anthropogenic sources emit a mixture of Hg^0 and ionic Hg in gas and particle forms.

The intercontinental transport of Hg has been observed in episodic events of elevated Hg^0 concentrations recorded at remote mountain top sites and during aircraft measurement campaigns. Such events observed in North America have been linked, based on backward trajectories and correlation with other atmospheric pollutant concentrations, such as co-emitted carbon monoxide (CO), to air masses originating over Asia. Analysis of such events suggests that Asian emissions have been underestimated in available emissions inventories.

Evidence for intercontinental transport of Hg into the Arctic, which has no primary anthropogenic sources, is also provided by observations of elevated levels of Hg in the tissue of Arctic wildlife.

Long-term changes in the atmospheric Hg burden have been derived from chemical analysis of lake sediments, ice cores, and peat deposits, and observed in firn air samples. Such evidence from both hemispheres suggests about a threefold increase of Hg deposition since pre-industrial times, emphasizing the importance of anthropogenic sources to current Hg levels in the environment. Measured deposition trends in Europe and North America are consistent with regional emission controls. However, global trends in concentrations and deposition are ambiguous, which may indicate off-setting effects between emission trends in Asia and the other parts of the world and significant recycling of Hg among environmental components. Our ability to understand these trends and the cycling of Hg is limited by the sparsity of long-term observations for Hg concentrations and deposition.

Persistent Organic Pollutants

POPs have long lifetimes in the environment, often cycling among different environmental compartments (i.e., air, water, soil, vegetation, snow, and ice). Thus, through direct emission and transport or repeated cycles of emission, transport, deposition and re-emission, POPs can end up in the environment far from their emission source. The overall potential and dominant mechanisms for intercontinental atmospheric transport vary among individual POPs, since these have widely different chemical characteristics. Evidence for intercontinental transport is provided from observations in remote locations far from emission sources and in elevated levels in plumes observed at mountain top

sites and during aircraft campaigns. Concentrations of POPs are often correlated with other anthropogenic pollutants.

Existing atmospheric monitoring programmes provide adequate spatial coverage of atmospheric concentration information for most POPs in the United Nations Economic Commission for Europe region, see Figure 1.3. The introduction of passive samplers that can measure air concentrations has significantly enhanced the spatial coverage of observations in other regions of the world. However, only a few monitoring programmes also analyse POPs in precipitation from which total deposition can be estimated.
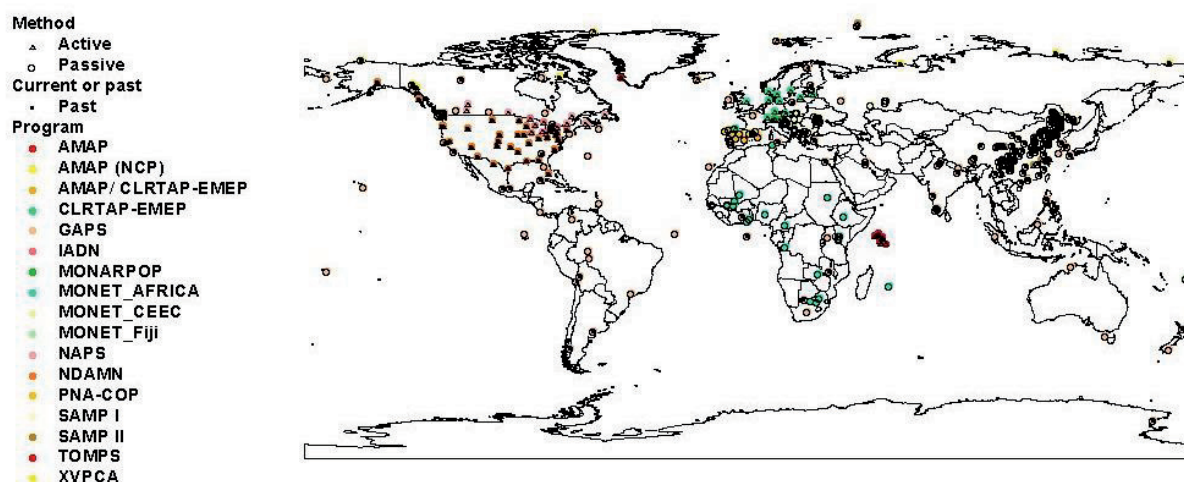
Figure 1.3. Atmospheric monitoring networks for POPs around the world.

Some long-term air monitoring programmes provide temporal trends with a time span of approximately 15 years, during which time international emission controls for many POPs have been implemented. For some POPs (e.g., technical Hexachlorocyclohexane (or HCH)), observed concentration trends show declines correlated with international emission controls. In other cases (e.g., p,p'- Dichlorodiphenyltrichloroethane (or DDT) at the Arctic stations of Alert and Pallas), observed trends show slow or no significant decline in air concentrations in response to international emission controls. The observed values have been shown to be correlated with climate change parameters, which affect the cycles of transport, deposition, and re-emission that drive intercontinental transport of these pollutants. However, our ability to understand the influence of these processes is limited by a lack of measurements and organized monitoring of POPs in media other than air.

Although the observational record is sufficient to demonstrate that PM, O_3, Hg, and POPs are transported on intercontinental scales, to understand the contribution of these intercontinental flows to health and environmental impacts and the significance of these flows for environmental management now and in the future, the observational evidence must be combined with quantitative models that describe the processes of emission, transport, transformation, and removal that drive the observed concentrations and deposition.

2. What are the main processes that drive these intercontinental flows and determine their magnitudes?

A Source Apportionment Construct

To understand the significance of the intercontinental transport of air pollution for air quality and other environment goals and the importance of the various processes which drive this transport, it is helpful to think about how air pollution concentrations and deposition may be apportioned back to emission sources. For O_3, PM, Hg, and POPs, observed concentrations or deposition at any given location can be thought of as composed of several different fractions, one of which is related to the intercontinental transport of anthropogenic emissions. Other fractions may be associated with natural emission sources or local and regional anthropogenic sources. Each fraction differs in terms of the emission sources that contribute to it, the temporal and spatial variability of the contribution, and the potential for and sensitivity to emission controls. Dividing observed concentrations and deposition into these distinct parts provides a construct to analyse the importance of different drivers and sources of air pollution now and in the future.

Attributing ground-level O_3 back to the emission sources that contribute to its formation is complicated by the fact that O_3 is not emitted, but is formed in the atmosphere through a non-linear system of photochemical reactions of emitted precursors, including nitrogen oxides (NO_X), volatile organic compounds (VOC), CO, and methane (CH_4). Observed surface O_3 concentrations may be thought of as composed of:

- O_3 that is transported from the stratosphere

- O_3 that is formed in the troposphere from

  - natural precursor emissions, including lightning, soil, fire, and vegetation emissions

  - anthropogenic precursor emissions that have been transported on intercontinental scales

  - anthropogenic precursor emissions from local or regional sources

The O_3 precursors from different sources interact in the atmosphere such that O_3 may be produced from natural or anthropogenic, local or transported, or a combination of natural and anthropogenic or local and transported emissions.

Each of the fractions that comprise observed O_3 differs in terms of (1) sensitivity to changes in precursor emissions, (2) the extent to which they can be controlled by a national or sub-national jurisdiction, and (3) the magnitude of the contribution. The relative contribution of each fraction varies widely by location and season and has evolved over time. We have confidence that human activities have contributed to the positive trend in mean surface O_3 levels observed in the Northern Hemisphere over the latter half of the twentieth century. However, there remain large uncertainties in our estimates of the source attribution for these changes.

PM concentrations and deposition are comprised of different chemical components, some of which are directly emitted as particles by natural or anthropogenic sources and are commonly referred to as primary PM. Other components are emitted by natural or anthropogenic sources as gases and form particles through chemical and physical reactions in the atmosphere and are commonly referred to as secondary PM. Both primary and secondary PM play a role in intercontinental transport. For purposes of understanding the role of intercontinental transport, PM concentrations or deposition may be thought of as composed of:

- volcanic eruptions, vegetation, and wind-blown dust (the last of which can be exacerbated by anthropogenic factors)

- open biomass (vegetation) burning, some of which is natural and some of which may be anthropogenic

- anthropogenic emissions that have been transported on intercontinental scales

- local and regional anthropogenic sources

The relative magnitudes of these fractions and their sensitivity to emission changes differ by location, season, year, and chemical component. Primary particles such as soil dust and black carbon, which are directly emitted into the atmosphere, respond linearly to changes in their emission sources. Secondary particles, such as sulphate and organic aerosols, are partly formed in the atmosphere through chemical and physical reactions of precursor gases that were directly emitted. These secondary particles may have a slight non-linear response to emission changes due to their dependence on oxidation and other reactions. The fractions also differ to the extent to which they can be controlled by a local or regional jurisdiction.

For Hg, adverse environmental and health impacts are associated with deposition and entry into aquatic and terrestrial food chains. While Hg^0 tends to dominate atmospheric loading and is the form in which Hg is transported long distances, Hg deposition is dominated by ionic Hg. Dry deposition and wet removal are both significant contributors to the total deposition of Hg. Once deposited, Hg may be re-emitted to the atmosphere. Thus, for purposes of apportioning deposition and subsequent impacts back to emission sources, Hg deposition can be divided into the following four fractions:

- Hg that was released originally from natural sources as Hg^0 and subsequently oxidized and deposited

- legacy Hg that was released from anthropogenic sources originally and deposited before being re-emitted as Hg^0 and subsequently oxidized and deposited again

- newly-released Hg that was released from anthropogenic sources primarily as Hg^0 and transported on intercontinental scales before being oxidized and deposited

- newly-released Hg that was released from local and regional anthropogenic sources primarily as ionic Hg and quickly deposited

Similar to Hg, many POPs cycle back and forth between the atmosphere and other environmental media in cycles of emission, transport, deposition, and re-emission. Unlike Hg, most POPs do not have significant natural sources, except for polycyclic aromatic hydrocarbons (PAHs) associated with natural biomass burning. Thus, observed POPs concentrations or deposition can be apportioned into the following fractions:

- re-emitted POPs that were originally released from anthropogenic sources then deposited before being re-emitted

- newly-released POPs that were released from anthropogenic sources and have been transported on intercontinental scales

- newly-released POPs that were released from local and regional anthropogenic sources

The relative importance of each of these fractions varies depending on the individual compound of interest and the location of the receptor relative to sources. POPs differ in terms of gas-particle partitioning, degradation pathways, and deposition processes, affecting the potential for atmospheric transport on relatively short time scales (monthly or annual basis). Some POPs may cycle between the atmosphere and other environmental compartments (i.e., water, soil, vegetation, snow, and ice) for decades and re-emission may be the largest source of input to the atmosphere. For very long-lived pollutants, transport in ocean currents also plays an important role in determining levels in locations remote from sources.

Drivers of Intercontinental Transport

The contribution that each of the fractions described above makes to the concentration or deposition observed at any given location is driven by the circulation of the atmosphere, the location and magnitude of emissions, chemical and physical transformations of the pollutants, and interactions

with and transformations in other environmental compartments. These processes are described in more detail below.

Atmospheric Circulation

In the mid-latitudes of the Northern Hemisphere, the general circulation is dominated by westerly winds that flow from Asia across the North Pacific Ocean to North America, from North America across the North Atlantic Ocean to Europe, and from Europe into Asia. These prevailing winds are depicted in Figure 2.1. Pollution is entrained in this flow by passing frontal systems and by deep convection that lift pollutants out of the lowest layer of the troposphere, known as the atmospheric boundary layer (ABL), and into the mid- and upper troposphere where it can be rapidly transported towards a downwind continent. Intercontinental transport also occurs in the boundary layer, but at slower speeds. Long-range transport of pollutants can produce distinct plumes in the mid- and upper troposphere, but to be relevant to air quality in a downwind continent, the pollution must descend to the surface. As the plumes descend they are diluted and can be difficult to distinguish from local pollution, especially in receptor regions with relatively high emissions.

The tropics and subtropics are dominated by the Hadley cell, with rising motion within deep convective cumulus towers in the deep tropics, which is balanced by subsiding motion in the subtropics. In the tropics, intercontinental transport is generally from east to west, guided by the trade winds throughout the lower and mid-troposphere.

In the Northern Hemisphere polar region, the cold and stable lower troposphere forms a dome over the Arctic, largely isolating the region from low latitude pollution that is emitted into warm air masses that ascend into the mid- and upper troposphere above the Arctic. Pollutant transport into the Arctic lower troposphere occurs preferentially from Europe when the outer regions of the Arctic dome pass over northern Europe, take up fresh emissions and then retreat back into the Arctic.
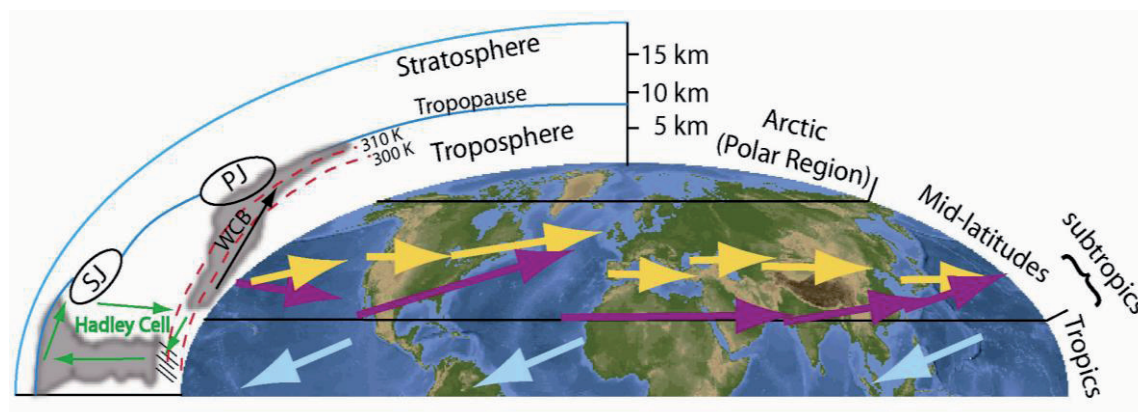
Figure 2.1. Schematic diagram showing some of the main features of the atmosphere related to the transport of air pollutants. The vertical cross section shows the average location of the tropopause (the boundary between the troposphere and stratosphere) and the polar (PJ) and subtropical (SJ) jet streams during winter. Vertical transport during winter is dominated by deep convective clouds in the tropics (the upward branch of the Hadley cell) and warm conveyor belts (WCBs) created by cyclones in the mid-latitudes. The average location of the jet stream is shown across the entire Northern Hemisphere for winter (magenta arrows) and summer (yellow arrows); locations of the tropical easterlies are also shown (light blue arrows). Also shown are the winter locations of the 300 K and 310 K potential temperature surfaces (red).

Emissions

The magnitude and impact of hemispheric and intercontinental scale transport of air pollutants is initially determined by the global distribution of emissions, and their spatial relation to the major meteorological transport pathways described above. Figure 2.2 shows the global distribution of anthropogenic NO_x emissions. Figure 2.3 shows the global distribution of

anthropogenic emissions of Hg, not including open biomass burning sources. The location of emissions with respect to the dominant atmospheric transport patterns has a strong influence on the frequency and strength of intercontinental pollution transport. For example, the intense emission regions along the east coasts of North America and Asia are at the origins of the North Atlantic and North Pacific mid-latitude cyclone storm tracks, which can loft the emissions and transport them to the free troposphere above downwind continents in a matter of days. With Western Europe located at the end of the North Atlantic storm track, its emissions are not lofted to the same extent as those on the east coasts of North America and Asia. Instead, European emissions are exported at relatively low altitudes and have a strong impact on the Arctic.
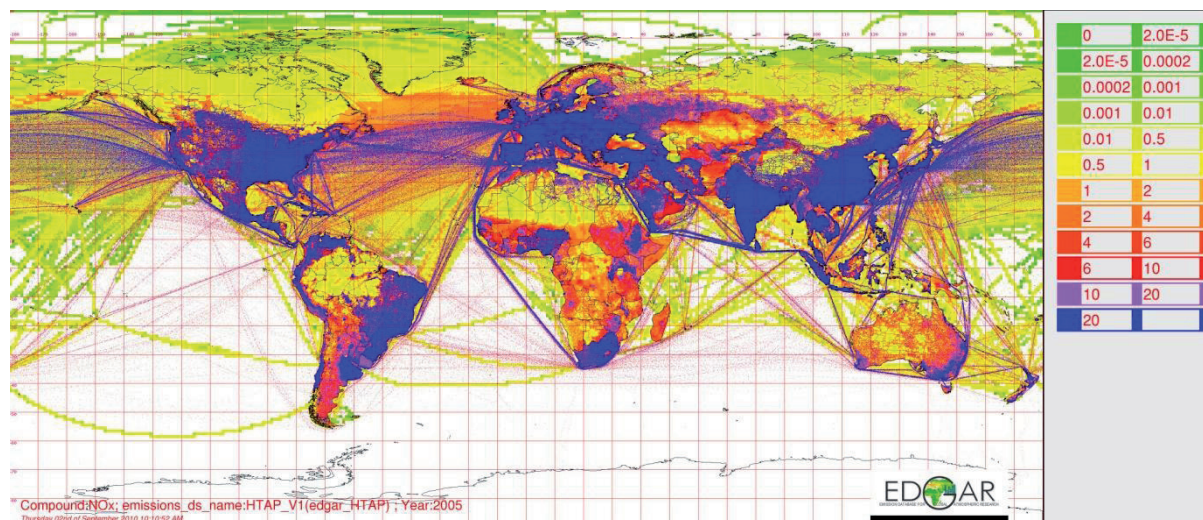
Figure 2.2. Gridded map of anthropogenic NO_x emissions in 2005 from EDGAR-HTAP emissions database on a $0.1° × 0.1°$ grid (in tons/grid cell). (No emissions from forest fires or savannah burning are included.) [Graphic courtesy of Emission Database for Global Atmospheric Research (EDGAR), European Commission Joint Research Centre and Netherlands Environmental Assessment Agency.]
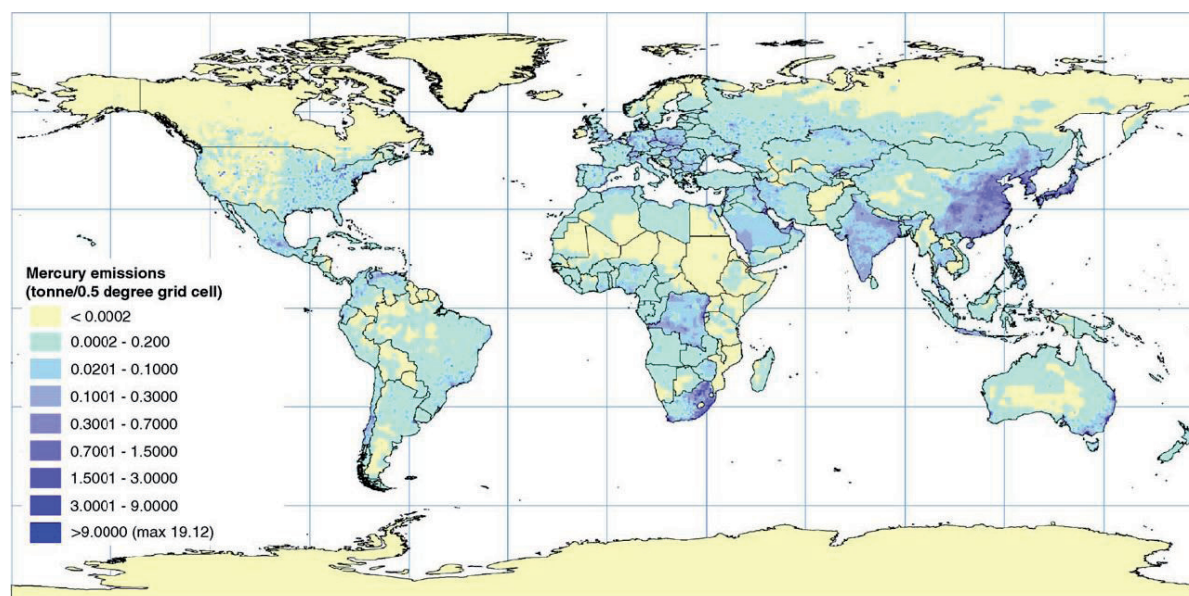
Figure 2.3. Spatial distribution of anthropogenic mercury emissions (combined point and 'distributed' sources for all species in all emission height classes) in 2000, within the Z05 (0.5x0.51) grid. [Reprinted from Figure 5 from Wilson, S. J., et al. (2006), Mapping the spatial distribution of global anthropogenic mercury atmospheric emission inventories, *Atmospheric Environment, 40*(24): 4621-4632, with permission from Elsevier.]

Many of the anthropogenic sources of O_3 and PM precursors and Hg emissions are associated with fossil fuel combustion or open biomass burning, and therefore, are similarly distributed. Many POPs, however, are pesticides or industrial chemicals that have very different patterns of manufacture and use. The manufacture or use of many of these chemicals is now banned in many countries. Thus, the importance of intercontinental transport is strongly dependent on where the chemicals have been used in the past or are still in use.

Chemical and Physical Transformations

Intercontinental pollution transport occurs on timescales of days to weeks, longer than the atmospheric lifetimes of some pollutants, and ample time for the trace gases and PM emitted or produced at the source to undergo removal or chemical transformation. By the time a polluted air mass arrives at a downwind continent, it is likely to have very different chemical properties than it did at the source. The chemical transformation processes that occur during intercontinental transport are numerous and complex.

Using transport from Asia to North America as an example, Figure 2.4 depicts a few of the most important chemical transformation processes for O_3 and PM that take place in a polluted air mass during transport. The chemical and physical transformation and removal processes that take place during transport differ depending on whether the polluted air is transported in the atmospheric boundary layer near the surface or the polluted air is lifted to the mid- and upper troposphere. These processes are described in more detail in Part A, Section 1.2.3.
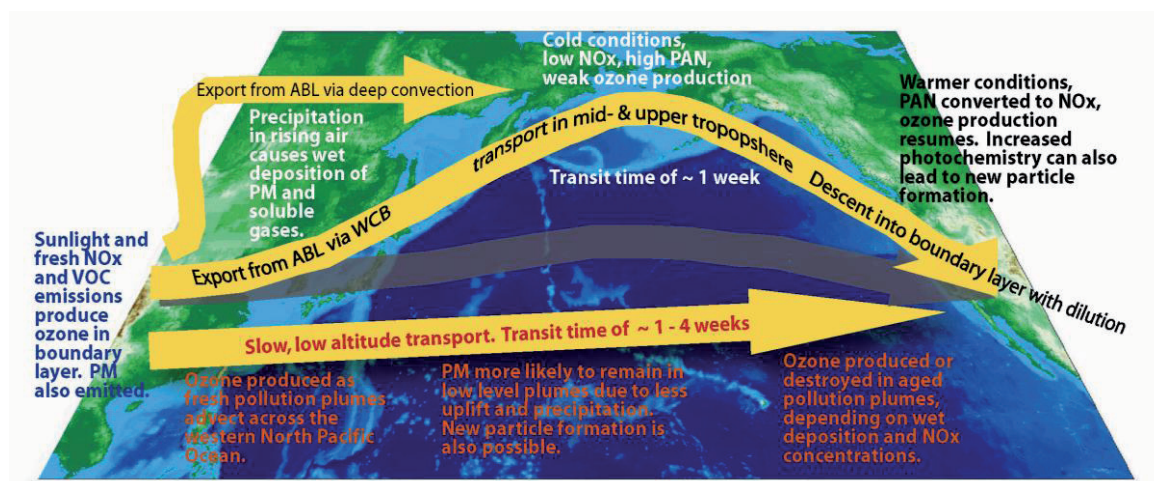
Figure 2.4. Transport and transformation processes during intercontinental transport of O_3 and PM. Blue text on the left applies to continental boundary layer processes, red text apples to low level transport and black/white text applies to high altitude transport.

For Hg, transformations between elemental and oxidized forms and the partitioning between the gas, aqueous, and particle phases largely determine whether Hg is transported long distances or is removed from the atmosphere. Hg is emitted into the atmosphere as Hg^0 in gaseous form or as oxidized, ionic Hg (Hg^{II}) in gaseous form or bound to suspended particulate matter (Hg^P). Hg^0, which has an average atmospheric lifetime of six months to a year, dominates atmospheric loading and is typically the form in which Hg is transported long distances. Ionic Hg is removed from the atmosphere very quickly through dry deposition and wet removal processes. These processes are illustrated in Figure 2.5.
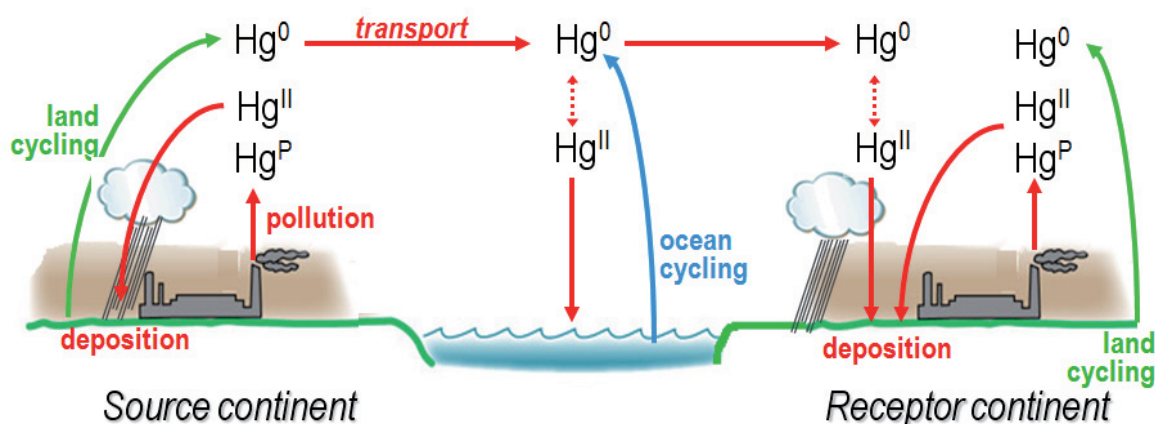
Figure 2.5. Transport and transformation processes during intercontinental transport of Hg. [Graphic courtesy of Lyatt Jaegle, University of Washington.]

For POPs, residence time in the atmosphere is largely determined by the rate of degradation through chemical reactions (primarily with hydroxyl radicals) and the rate of wet or dry deposition to surfaces. Many POPs are semi-volatile, such that they distribute between the gas phase and aerosols in the atmosphere, affecting the rates of degradation and deposition. Once deposited, semi-volatile POPs may also be re-volatilized, re-entering the atmosphere. Through such re-emission, some POPs are able to be transported very long distances in multiple "hops" consisting of repeated cycles of transport, deposition, and volatilization.

Interactions With and Transformations In Other Environmental Compartments

Because of the cycling of Hg and POPs among the atmosphere and land, ocean, and vegetative surfaces, models of the atmospheric transport of Hg and POPs must account for the emission or re-emission of Hg and POPs from surfaces and the chemical and physical processes that occur in surface media that permanently remove the substances from circulation in the environment. Figure 2.6 depicts the various environmental compartments and the linkages among them that must be taken into account to understand Hg and POPs transport.

Studies using spatially and temporally resolved transport models show that substances do not have inherent total environmental lifetimes and long-range transport potentials are not intrinsic substance properties. The potential for long-range transport depends strongly on the time, location, chemical and physical form of emissions; the concentration of other substances in the atmosphere and other environmental media; and the past history of emissions and environmental conditions. For example, the mean total environmental lifetime of DDT applied in agriculture in the 1980s was considerably longer than in the 1970s and 1990s, in large part due to shifts in the geographic distribution of DDT use that occurred over this period.

Air-sea exchange of semi-volatile POPs and Hg allows for multiple cycles through the atmosphere and ocean. Ocean transport is expected to be most important for persistent substances that are highly water soluble and have low vapour pressure, such as perfluorinated acids, which are a relatively new class of POPs. In the Northern Hemisphere, atmospheric transport tends to be zonal (west-east) in nature, whereas oceanic transport enhances meridional (north-south) transport as relatively fast currents exist along the African and American continents, leading to net northward transport in the Northern Hemisphere oceans. On the other hand, transfer of contaminants from surface waters to the deep sea by both deep water formation and sinking with sedimenting particulate matter may slow down long-range transport and suppress the mobility of pollutants.

Moreover, the public health and environmental impacts associated with Hg and POPs pollution are realized primarily through the contamination of aquatic and terrestrial food webs. To assess these impacts, one must take into account the chemical and physical transformations that occur in other media (e.g., methylation of Hg), as well as processes of bioconcentration, bioaccumulation, and biomagnification in food webs.
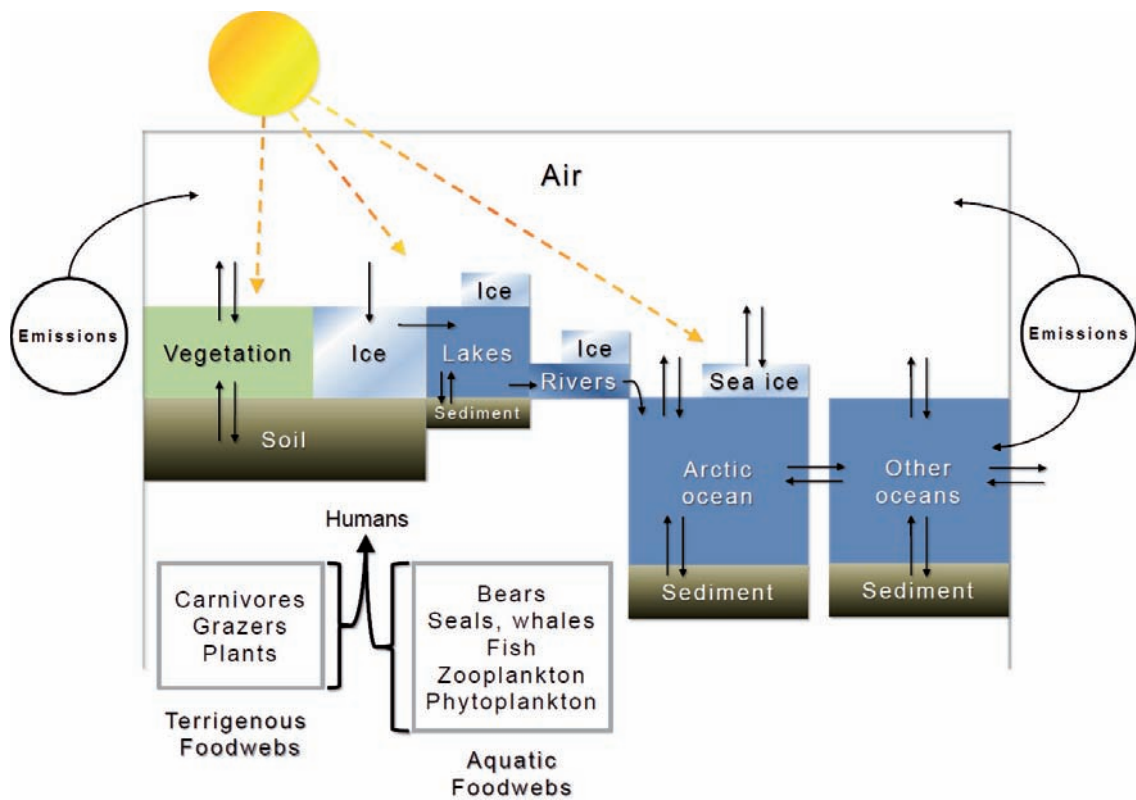
Figure 2.6. Examples of linkages among environmental compartments that must be taken into account to understand impacts of Hg and POPs pollution transport.

3. **What do current models tell us about the contribution of intercontinental or global flows to concentrations and deposition in the different regions of the Northern Hemisphere and how changes in current emissions in one region affect air pollution concentrations or deposition in another region?**

Using current quantitative models, in conjunction with emission estimates and observations, the contribution of intercontinental transport to concentrations and deposition in different regions of the world may be estimated. These estimates may be based on methods that "tag" emissions and trace them through the model from emission to concentration or deposition. Alternatively, these estimates may be based on the sensitivity of concentrations or deposition to changes in emissions. In the case of a largely inert pollutant that is permanently removed from the atmosphere upon deposition, concentrations are linearly related to emissions and these two modelling approaches give similar results as the contribution of all sources at a particular receptor is additive. This linear assumption is a good approximation for most PM components and the newly-emitted fractions of Hg and POPs. However, for O_3, which has a non-linear dependence on precursor emissions, and re-emissions of Hg and POPs, which are dependent on past emission histories and emissions from other media, the two methods may give different perceptions of the importance of different sources.

To characterize the magnitude of intercontinental transport and the level of agreement among current models, the TF HTAP organized a series of multi-model experiments. The first set of these experiments was based on the emissions sensitivity approach, examining the impacts of 20% emission decreases of relevant anthropogenic pollutants in four regions, approximately covering the major populated areas of North America, Europe, South Asia, and East Asia. These regions are shown in Figure 3.1. These names are used hereinafter to refer to these rectangular regions, which encompass more than 75% of the anthropogenic emission sources in the Northern Hemisphere but also include significant areas of ocean. Specific analyses were also made to quantify the impact in the Arctic of emission changes in these four source regions. The findings from these experiments for each of the pollutants of interest are discussed below. The limitations of the models are discussed in more detail in the response to Question 6.
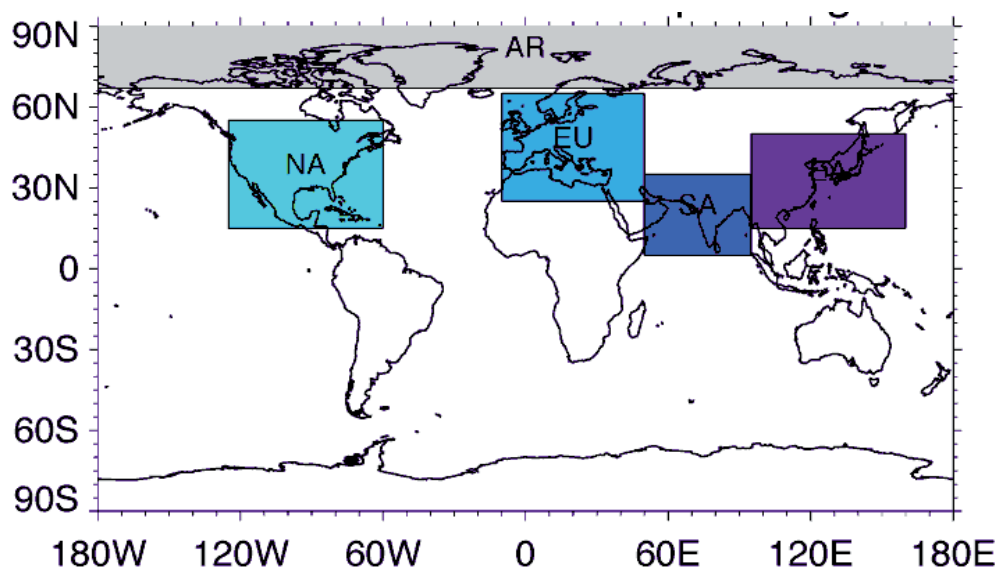
Figure 3.1. The main source and receptor regions used in the HTAP multi-model experiments: North America (NA), Europe (EU), South Asia (SA), and East Asia (EA). Additional analyses examined the Arctic (AR) as a receptor region. For PM and Hg, some additional source and receptor regions relevant to those issues were examined.

To quantify the relative importance of emission changes outside each of these regions, as compared to emission changes inside each of these regions, we defined the Relative Annual Intercontinental Response (RAIR) metric. RAIR is defined as the sum of the changes in the annual, regionally-averaged concentration within a region due to a 20% decrease in emissions in the three other regions divided by the sum of the changes in concentration within a region due to a 20% decrease in emissions in all four regions. The value of the metric ranges from 0%, indicating no intercontinental influence, up to 100%, indicating that air quality in a region is completely dominated by intercontinental sources. Thus, the RAIR is a measure of how much benefit a region may receive from emission reductions in other regions when emission reductions are coordinated on an intercontinental scale.

Ozone

The annual average ground-level O_3 mixing ratio averaged across the four regions and the ensemble of participating models is about 37 parts per billion by volume (ppbv) ($\pm$ 4 ppbv standard deviation). This annual average, region-wide O_3 concentration masks large seasonal and geographic variability, and large differences across models. However, it provides a useful point of reference for considering the magnitude of intercontinental transport. Based on estimates in the literature, about 20-25% of this annual average ground-level concentration originates in the stratosphere and a similar fraction is formed from natural emissions of precursors. The remainder is due to anthropogenic sources of precursors from within the region itself and transported from outside the region. The relative contribution of anthropogenic and natural, regional and extra-regional sources varies by location, season, and year.

Models indicate that for Europe, East Asia, and North America, the absolute contribution of intercontinental transport to ground-level O_3 typically peaks in spring and fall, and is smallest during summer when O_3 concentrations reach their highest maxima due to the peak in production from local and regional emissions. In these regions in winter, intercontinental transport of O_3 may exceed local and regional production. For South Asia, which is largely dominated by the summer and winter monsoon regimes, the absolute contribution of both intercontinental transport and local and regional production of O_3 are largest during the winter monsoon period and the annual contribution of intercontinental transport does not exceed that of local and regional production. Intercontinental contributions to ground-level O_3 in the Arctic are larger than production from local and regional emissions and peak in April through June, with a secondary maximum in October and November.

Model calculations find that the maximum influence of intercontinental transport on ground-level O_3 generally occurs on mid-range pollution days (i.e., near the middle of the O_3 probability distribution, typically 50-70 ppbv), suggesting that import is less significant on days with the highest O_3 levels. These high O_3 conditions are typically due to trapping of local precursor emissions under stagnant meteorological conditions, which also suppress the influence of distant sources. However, there are locations and times that intercontinental transport makes substantial, even dominant, contributions to high surface O_3 episodes. Overall, the results indicate that decreasing local or regional emissions is generally more effective at decreasing the highest O_3 levels, but that O_3 associated with stratospheric origins, natural emissions sources, and intercontinental transport comprise a significant fraction of tropospheric O_3 that is not within the control of local and regional political jurisdictions.

For O_3, the impact of 20% changes in anthropogenic emissions (of NO_X, VOC, CO, sulphur dioxide (SO_2), and direct PM) in one region on surface O_3 in the other regions varies from 0.07 to 0.37 ppbv on an annual average, region-wide basis, as estimated by the mean of the model ensemble (see Table 3.1.). These values are significant in comparison to the response of surface O_3 to 20% decreases of emissions within the region itself, which vary from 0.8 to 1.3 ppbv. These annual average, region-wide values mask large temporal and geographic variability. For example, the values can vary by up to a factor of two from month to month and from grid cell to grid cell within a region.

Table 3.1. Annual and spatial mean surface O_3 response (ppbv) to 20% decreases in anthropogenic precursor emissions (NO_x, CO, VOC, plus aerosols and their precursors). Values are mean ± one standard deviation across the 15 models that conducted the regional perturbation simulations. Bold font denotes responses to foreign emission perturbations that are at least 10% of the response to domestic emission perturbations. Also shown is the relative annual intercontinental response for each receptor region defined as the ratio of the total response in mean surface O_3 due to changes in the other three source regions compared to that due to changes in all regions. (NA: North America, EU: Europe, EA: East Asia, SA: South Asia)

Source Region	Receptor Region			
	NA	EU	EA	SA
Annual mean decrease				
NA	1.04±0.23	**0.37±0.10**	**0.22±0.05**	**0.17±0.04**
EU	**0.19±0.06**	0.82±0.29	**0.24±0.08**	**0.24±0.05**
EA	**0.22±0.06**	**0.17±0.05**	0.91±0.23	**0.17±0.05**
SA	0.07±0.03	0.07±0.03	**0.14±0.03**	1.26±0.26
Relative Annual Intercontinental Response (RAIR)				
	32%	43%	40%	32%

The largest source-receptor relationship, in an absolute sense, is the impact of North American emissions on European surface O_3 levels. This is followed by the impact of European emissions on South Asian and East Asian surface O_3. The annual average impact of East Asian emissions on North American surface O_3 is similar to the impact of North American emissions on East Asian surface O_3, but with peaks in different seasons. European emissions have the largest influence on Arctic surface O_3.

The RAIRs for ground-level O_3 range from 43% for Europe, to 40% for East Asia, to 32% for South Asia and North America. These values suggest that, in all four of the source-receptor regions, at least 30% of the total concentration changes within each of the regions is related to emission changes in the other three regions. Substantial O_3 transport takes place above the boundary layer in the free troposphere, where it can be brought to lower altitudes over distant receptors during subsidence, and mix with local emissions. The RAIRs are larger for column O_3 than for ground-level O_3, and the column RAIRs exceed 50% in Europe and East Asia.

Imported O_3 and precursors may have qualitatively different impacts on urban areas than rural areas due to the strong dependency of O_3 chemistry on the ratio of NO_X to VOCs. Results from the global models used in the HTAP multi-model experiments suggest that the response of annual average O_3 concentrations in large cities to changes in intercontinental transport may be as large or larger than the response to changes in regional emissions. However, global models are not well suited to estimate responses at urban scales, as the resolution of current models is too coarse to resolve the strong chemical contrasts associated with urban regions. New studies using regional air quality models show that the higher resolution models are better able to resolve local topography, finer variations in land cover and use, and O_3-VOC-NO_X chemical non-linearities, which all have effects on the strength of surface O_3 responses to emission changes. For example in East Asia, results using a regional model predicted a smaller mean response in O_3 from European sources, but with much larger spatial variability.

The HTAP multi-model experiments also examined the sensitivity of O_3 to changes in CH_4 by decreasing the globally fixed CH_4 concentration by 20%. From those simulations, we estimated the response to 20% decreases in regional anthropogenic CH_4 emissions. While local and regional emission controls are clearly most effective for lowering local and regional O_3, the O_3 response to anthropogenic emissions of CH_4 from distant source regions is nearly as large as that to emissions of

the traditional O_3 precursors in these regions. The O_3 response to changes in CH_4 emissions requires several decades to be fully realized, given the relatively long atmospheric lifetime of CH_4. Thus, controlling CH_4 is an important component of a strategy to limit increases in baseline surface O_3, and as an important greenhouse gas, has additional benefits for climate change mitigation.

Particulate Matter

There is a wide range of surface aerosol concentrations predicted by current models, reflecting large uncertainties in emissions and atmospheric processes as represented in the models. The HTAP multi-model experiments suggest that, in the four regions studied, ground-level concentrations of wind-blown soil dust from deserts in Africa, Asia, and the Middle East can be a factor of 1.5 to almost 20 higher than that from anthropogenic and open biomass burning sources on a region-wide, annual average basis. The large contribution of wind–blown soil dust to the regional average concentrations is due in large part to the fact that the boundaries of the four study regions were drawn in a manner that encompasses large areas of desert in North Africa, Asia, and the Middle East. Of the ground-level PM concentration originating from anthropogenic and open biomass burning sources, intercontinental transport of anthropogenic emissions accounts for between 5% and 35% and transport from open biomass burning contributes between 4% and 14% on a region-wide, annual-average basis in each region.

In terms of the chemical components of PM, the HTAP multi-model experiments provided information for four components: sulphate, black carbon, particulate organic matter (POM), and mineral (or soil) dust. For North America, Europe, South Asia, and East Asia, anthropogenic sources of emissions within each region account for 60-90% of the sulphate and black carbon concentrations. For POM, the contribution of anthropogenic sources within the region is less than for sulphate and black carbon, with biomass burning and biogenic emissions also contributing to the estimated concentrations.

In the Arctic, European pollution is the largest contributor to surface sulphate, followed by volcanic emissions. Biomass burning in the boreal forests of Eurasia plays a major role in determining the surface concentrations of black carbon and POM in the Arctic. Nearly half of the mineral dust at the Arctic surface is from Asia, with smaller amounts from Africa and the Middle East.

The fraction of PM associated with intercontinental transport increases with altitude, indicating the importance of transport above the boundary layer. As a result, the contribution of intercontinental transport to total column loading tends to be 15 to 25% higher than the contribution to surface concentrations or deposition.

In many regions deposition of oxidised sulphur and nitrogen is large, and exceedance of critical loads occurs widely over many ecosystems. The intercontinental contribution to these exceedances is small. However, particular attention should be paid to the effects of long range transport on sensitive ecosystems.

As is the case with O_3, annual average, region-wide statistics mask significant variability among seasons and within a given region. The seasonal cycles of aerosol concentration and intercontinental transport vary by chemical component and by region, and there are substantial differences among the seasonal cycles in surface concentrations predicted by models participating in the HTAP multi-model experiments. The model differences are larger for dust than they are for sulphate, black carbon, and POM, and are larger for the Arctic than for the mid-latitude regions, reflecting large uncertainties in knowledge of sources, transport, and removal processes related to the Arctic. However, despite the substantial differences in estimated absolute concentrations, there is much closer agreement among the participating models in terms of the source-receptor relationships on an annual average, region-wide basis.

As presented in Table 3.2, the RAIRs estimated in the HTAP multi-model experiments indicate that decreases in intercontinental transport would contribute 5% to 20% of the decrease in region-wide, annual average, ground level PM concentrations that results from decreasing anthropogenic emissions by 20% in each of the four regions studied. The RAIRs for surface deposition of sulphate, reactive nitrogen, black carbon and POM are similar to those for surface concentrations. The RAIRs for aerosol

column loadings are generally larger (24%-37% for sulphate, 15%-24% for black carbon, and 12% to 23% for POM), reflecting the importance of transport above the boundary layer and implying more significant contributions to visibility and radiative forcing impacts.

Ground-level PM concentrations and deposition in the Arctic are most sensitive to emission changes in Europe. However, total column loadings of PM over the Arctic are equally sensitive to changes in emissions from Europe or Asia, due to the fact that Asian emissions have a stronger tendency to be lifted and transported aloft than do European emissions.

Ground-level PM concentrations generally respond linearly to changing emissions from both local and upwind source regions. However, sulphate and some fraction of POM are not directly emitted, but are formed in the atmosphere through oxidation, and can be affected by non-linear systems of chemical reactions. For example, present-day North American emissions of NO_X and CO, together, are estimated to increase surface PM in Europe and East Asia by up to 0.5 micrograms per cubic metre ($\mu g/m^3$). These impacts are not due to direct transport of secondary PM produced in the source region and then transported to other continents, which is negligibly small. Rather, the increases are due to increases in oxidants, leading to enhanced production rates of secondary aerosols during long range transport and in the receptor region. These results underscore the multifaceted nature of the consequences of a rising background of pollution levels in the Northern Hemisphere on local pollution levels.

Table 3.2. The Relative Annual Intercontinental Response (RAIR) of pollutants to 20% emission decreases in four regions approximating North America, Europe, South Asia, and East Asia. The RAIR is the sum of the changes in the annual, regionally-averaged concentration within a region due to a 20% decrease in emissions in the three other regions divided by the sum of the changes in concentration within a region due to a 20% decrease in emissions in all four regions. Thus, the RAIR is a measure of how much benefit a region may receive from emission reductions in other regions when emission reductions are coordinated on an intercontinental scale.

Scenario	Pollutant/Parameter[a]	Receptor Region			
		North America	Europe	South Asia	East Asia
2001	O$_3$ Concentration	32%	43%	32%	40%
2001	O$_3$ Total Column Burden	39%	62%	38%	60%
2001	PM Concentration[b]	7%	5%	20%	9%
2001	SO$_4$ Deposition	8%	9%	24%	12%
2001	Black Carbon Deposition	4%	1%	12%	3%
2001	Reactive Nitrogen Deposition	3%	4%	13%	7%
2001	Aerosol Optical Depth[c]	17%	13%	25%	17%
2001	SO$_4$ Total Column Burden	25%	25%	37%	24%
2001	Hg Deposition	61%	35%	43%	10%
2001	α-HCH Deposition	82%	9%	6%	63%
2001	PCB-28 Deposition	9%	3%	48%	56%
2001	PCB-153 Deposition	11%	2%	32%	50%
2001	PCB-180 Deposition	11%	4%	36%	56%

Notes:
[a] Concentrations are at ground-level.
[b] Estimated as the sum of sulphate and particulate organic matter.
[c] Aerosol Optical Depth calculated for the sum of sulphate, black carbon, and particulate organic matter. Soil dust was not included.

Mercury

Based on the HTAP multi-model experiments, natural and re-emitted Hg account for about 35% to 70% of total Hg deposition on a region-wide, annual average basis depending on the region, whereas intercontinental transport of newly-released anthropogenic Hg emissions accounts for about

10% to 30% of total Hg deposition, on a region-wide, annual average basis. East Asia, which accounted for almost 40% of total global newly-released Hg in 2000, is the most dominant among the four HTAP source regions, accounting for 10% to 14% of the annual Hg deposition found in other regions, followed by contributions from Europe, South Asia, and North America. However, where deposition is highest, local and regional anthropogenic emission sources are the dominant sources of Hg deposition.

Although no seasonal changes in anthropogenic emissions were considered in the models, strong seasonal variation in simulated total deposition was found. This can be attributed to changes in Hg^0 oxidation chemistry, seasonal cycles in meteorological and land cover parameters (atmospheric stability, precipitation, and vegetation canopy) and regional patterns of atmospheric transport.

The Arctic has no anthropogenic emission sources within the region, so anthropogenic Hg that is deposited there comes from intercontinental transport. A large fraction of transported Hg is deposited in springtime during polar sunrise in atmospheric Hg depletion events (AMDEs), during which Hg^0 is rapidly oxidized by photochemical reactants that have built up over the dark winter. Current models show significant deviation in estimates of Hg deposition to the Arctic due to the uncertainties in the model formulation of the processes related to AMDEs and transport to the polar region.

The global Hg models participating in the HTAP multi-model experiments provided relatively consistent estimates of the impact of one source region on another despite significant differences in emissions and chemistry in each model. In these global models, the change in Hg deposition in one region is linearly related to the change in emissions in another region, provided that the relative proportion of Hg^0 and ionic Hg in the anthropogenic emissions stays the same in both regions. Increases in Hg^0 emissions lead to enhanced export, whereas increases in ionic Hg result in higher local deposition in the region itself.

Given that emissions of Hg are not equally distributed among the HTAP source regions, the overall influence of the different regions on one another is not similar. The multi-model mean RAIRs shown in Table 3.2 vary from a low of 10% for East Asia to a high around 60% for North America. These results suggest that intercontinental transport is not a significant influence on Hg deposition in East Asia, but intercontinental transport is a significant source of deposition in North America, South Asia, and Europe. Furthermore, the results suggest that similar percentage emission reductions in other parts of the world will decrease deposition in North America more than the same percentage emission reductions in North America itself.

In the Arctic, Hg deposition could be most efficiently controlled by emission reductions in East Asia and Europe due to their proximity to the Arctic, prevailing atmospheric circulation patterns, and the significant contribution of these regions to global anthropogenic emissions.

Within a receptor region, proximity to the intercontinental source region has a small effect on the spatial distribution of Hg deposition. For example, relative Hg deposition decrease in North America caused by emission reduction in East Asia is somewhat higher in the western part of North America, whereas response to the European emission reduction prevails in eastern North America.

The response of Hg deposition to changes in emissions varies with season. Intercontinental transport may be a factor of two more important in summer than in winter due to more active mixing of the boundary layer with air aloft. Thus, seasonal variation of the deposition response to emission reduction in other continents can reach 30% of the annual mean. Distinctly different seasonal cycles characterize the transport between adjacent regions such as East Asia and South Asia or Europe and South Asia, which are driven by changes in monsoonal systems. For example, the most significant effect of South Asian emission reduction on Hg deposition in East Asia takes place during the first half of a year, whereas the opposite is true of the effect of East Asian sources on South Asia, which is most significant in the second half. Similarly, South Asian sources have a somewhat larger influence on Europe in winter; whereas European sources more strongly affect Hg deposition to South Asia in summer.

In the year-long simulations, the large contribution of natural sources and re-emitted legacy Hg to deposition dampens the relative response of Hg deposition to the reduction in new anthropogenic emissions, whether associated with local and regional sources or intercontinental transport. If integrated over much longer time scales, the decrease in deposition will be larger as decreasing new emissions will slowly decrease the amount of legacy Hg that is cycling in the environment.

Persistent Organic Pollutants

As part of the HTAP multi-model experiments, the transport of several POPs (Polychlorinated biphenyl-28 (PCB-28), PCB-153, PCB-180, and α-HCH), covering a range of different physical and chemical properties and for which some emissions and observational data are available, was simulated by three models. The three models used different modelling approaches to describe pollutant transport through environmental compartments and different spatial resolutions. Despite their differences, the models provide generally consistent estimates of annual average atmospheric concentrations, the major transport pathways of the selected POPs, and responses to primary emission changes. In most of the cases, the differences between the mean response and the model estimates are within a factor of 2 to 3.

As with the other pollutants, the largest impact on deposition, in most cases, results from changes in local and regional emissions within the region, especially for regions with higher emissions (North America and Europe for PCBs, and South Asia and Europe for α-HCH).

Differences in the estimated RAIRs for POPs presented in Table 3.2 are primarily a function of the uneven spatial distribution of emissions among regions and the transport characteristics of the pollutant. The largest effect of intercontinental transport is seen for α-HCH deposition in North America and East Asia, 82% and 63%, respectively, due to their relatively low contributions to the global emissions. For Europe and South Asia, the RAIR values are quite low (9% and 6%), indicating that the decrease of their emissions is more important for the reduction of pollution levels rather than changes of emissions in foreign regions. For PCBs the largest sensitivity to intercontinental transport is obtained for South and East Asia, varying from 32% to 56%. Levels of PCB deposition in Europe and North America are influenced largely by the changes in their own emissions.

Arctic pollution is mostly sensitive to the changes of emissions in Europe (for all simulated POPs) followed by North America (for PCBs) and South Asia (for α-HCH).

The regional differences in the efficiency of atmospheric POPs transport alone were explored by simulating the transport of selected POPs assuming an equal mass of emissions in each source region. In contrast to the results with regionally different emissions, the uniform emissions produced the same level of response to emission changes for all source-receptor region pairs, about 1.5%-2%. A slightly more significant effect is obtained for the closest pair of regions, South Asia's influence on East Asia (2-3%). From the equal emission simulations, transport to the Arctic is most efficient from Europe, producing a 5-6% response. A slightly lower response in the Arctic can be seen for emission changes in North America and East Asia (about 4%) and the Arctic response is weakest for South Asian emissions (about 2%).

4. What is the contribution of these intercontinental or global flows to impacts on human health, natural and agricultural ecosystems, and near-term climate change?

The intercontinental transport of O_3, PM, Hg, and POPs contribute to serious public health problems and damage to natural and agricultural ecosystems in many parts of the world. O_3 and PM also contribute significantly to climate change on regional and global scales. Each of these types of impacts is discussed further below.

Ozone

There is considerable evidence from experimental human and animal studies and epidemiological studies that exposure to ambient O_3 concentrations causes adverse health effects which range from minor sensory irritation to premature death.

The highest concentrations of O_3 are typically associated with stagnant conditions, when the contribution from intercontinental transport of air pollution is low and the contribution of local and regional sources are most important. However, intercontinental transport has increased baseline O_3 concentrations to the point where they exceed thresholds for protection of vegetation in many locations and exceed thresholds for the protection of human health occasionally in some locations. As public health-based air quality standards continue to be tightened based on new health effects research, the contribution of intercontinental transport to concentrations that exceed such standards will continue to increase.

Relatively few studies have tried to quantify the human health impacts of intercontinental transport of O_3 specifically. Those studies have focused on the relationship between annual average concentrations and premature mortality and suggest that intercontinental transport can contribute significantly to health impacts of air pollution within a given receptor region. One study based on the HTAP multi-model experiments estimated that intercontinental transport of O_3 contributes from 20% to more than 50% of O_3-related premature adult mortalities in a given receptor region, subject to large uncertainty.

The sum of the health impacts of transported pollution in downwind foreign regions can be larger than the health impacts of emissions in the source region itself. Although the impact on ambient concentrations in downwind foreign regions may be much less than in the source region itself, the total population exposed in those downwind regions is much greater. The HTAP multi-model experiments suggest that emission reductions in North America and Europe will avoid more O_3-related mortality outside these source regions than within the regions themselves.

O_3 causes damage to crops, forests, and grasslands, which has important implications for productivity, biodiversity, and food security. Recent experimental studies on field crops, adult forest stands and different grassland ecosystems have found significant impacts associated with ecologically realistic free-air O_3 fumigations that mimic the observed increases in baseline O_3 concentrations.

Global crop yield losses of four staple crops due to exposure to O_3 are estimated to be between 3% and 16%, depending on the crop, and are valued at $14 billion - $26 billion per year. Based on the HTAP multi-model experiments, intercontinental transport may be responsible for about 5% to 35% of the estimated crop yield losses depending on the location, crop, and response function used. However, there is significant uncertainty in these estimates, part of which is due to the limited representativeness of available exposure-response functions based on threshold indices (e.g., accumulated ozone exposure over a threshold of 40 parts per billion (AOT40) and the sum of all hourly ozone concentrations greater than 0.06 parts per million (SUM06)).

O_3 contributes significantly to climate forcing, directly as a greenhouse gas and indirectly by damaging plants and inhibiting their natural uptake of carbon dioxide (CO_2). Among O_3 precursors, widespread decreases in emissions of CH_4, CO, and VOCs will decrease net climate forcing. Decreasing NO_x may increase climate warming over decadal time scales because less NO_x leads to less hydroxyl radical, increasing the lifetime of CH_4, which is a greenhouse gas itself. Over time, the

increase in radiative forcing from the increased lifetime of CH_4 is greater than the decrease in radiative forcing from decreased O_3 formation. Decreasing emissions of CH_4, however, will result in decreases in the direct forcing from CH_4 and the direct and indirect forcing of O_3, affecting the rate of climate change in the coming decades.

The radiative forcing exerted by O_3 is not globally uniform, but extends from the location of precursor emissions over regional and intercontinental scales. This inhomogeneous forcing affects climate change at the global scale and at the regional scale, influencing atmospheric heating and dynamics and ultimately patterns of temperature and precipitation. The largest climatic impacts do not necessarily occur where the radiative forcing occurs and may occur downwind of the source region.

Particulate Matter

For PM, the experimental and epidemiological evidence for effects on mortality is stronger than it is for O_3. Although the highest PM concentrations are typically associated with local and regional emission sources, intercontinental transport events associated with forest fires or dust storms do produce concentrations that exceed short-term public health standards (e.g., the impact of Saharan dust events on PM levels in southern Europe). On a longer-term basis, current levels of intercontinental transport of PM interfere with the ability to meet visibility targets for natural surroundings in western North America. Intercontinental transport of PM components other than wind-blown dust or from fires is not usually sufficient to exceed health-based ambient standards.

Only a few studies have tried to quantify the human health impacts of intercontinental transport of fine particles specifically. Those studies conclude that contributions to PM from emissions within a region are expected to be much more important for human health than emissions from intercontinental transport. However, the impacts of transported PM are still significant. Based on the HTAP multi-model experiments, the intercontinental transport of PM has influences on human mortality that are comparable to O_3. While O_3 is transported among regions more efficiently, the relationship between PM and mortality is stronger. Consequently, the estimated mortalities attributable to PM within each source region are much higher, and the contributions of the three foreign regions to the mortality in a given home region range from 3 to 5%. Of the total mortalities associated with emissions from North America and Europe, 15% and 12%, respectively, are estimated to be realized outside of these source regions.

PM deposition also damages a variety of different ecosystems including forests and grasslands through acidification. Similarly, eutrophication is damaging to biodiversity in sensitive ecosystems of low nutrient status. PM can benefit ecosystems by increasing the diffuse solar radiation; however, the contribution of long-range transport to these effects is very uncertain and may be relatively small.

PM is a significant contributor to climate forcing; intercontinental transport influences the distributions of PM and, therefore, the extent and magnitude of its forcing. PM is a mixture containing components that mainly cool, including sulphate and organic aerosols, and black carbon that warms. Anthropogenic emissions of black carbon, CH_4, CO, and VOCs are estimated to have caused a climate forcing since 1750 roughly as large as that from anthropogenic CO_2. Reductions in PM would improve air quality, but for cooling aerosols, including sulphate, nitrate and POM, this would generally increase warming. Reductions in black carbon would typically benefit both air quality and climate.

As with O_3, the climate forcing resulting from changes in emissions of PM and its precursors depends strongly on location, timing, and the background composition. Based on the HTAP multi-model experiments, the RAIR of aerosol optical depth and direct radiative forcing was calculated for each of the four continental regions and each PM component. For all regions and PM components, emission changes outside the region had a significant effect relative to emission changes within the region, with RAIR of radiative forcing ranging from 9% to 30%. The change in radiative forcing over South Asia is most influenced by the import of sulphate aerosol, and North America is most influenced by the import of black carbon. For the change in radiative forcing due to sulphate, black carbon, and POM combined, South Asia is the region most strongly influenced by foreign sources,

and North America and Europe are the least. The variability among models is very large, highlighting the significant uncertainties in modelling aerosol processes and transport.

The Arctic is experiencing rapid climate change. Arctic climate is affected by PM and O_3 that are transported into the Arctic from other regions, as well as by the climate forcing of PM and O_3 outside of the Arctic. Deposition of black carbon on snow and ice is understood to be an important positive (warming) forcing in the Arctic.

Mercury

Hg differs from other major atmospheric pollutants (e.g., O_3 and PM) in that its environmental and health impacts are not directly related to its atmospheric burden. While the major redistribution of Hg is via the atmosphere, its primary environmental and health impact is in aquatic systems and for aquatic organisms and their consumers. Atmospheric Hg that is deposited directly or indirectly into aquatic systems is converted from an inorganic form to methylmercury (MeHg) by microbes in the water and sediments of wetlands, lakes, reservoirs, rivers, estuaries and oceans. Unlike other forms of Hg, MeHg biomagnifies in aquatic food webs. Consumption of fish or other aquatic organisms with elevated MeHg concentrations is the primary route of exposure for humans and wildlife.

Levels of MeHg in fish are used as the major environmental impact indicator of Hg contamination, and they respond both to changes in atmospheric Hg inputs and composition and changes in environmental conditions in the atmosphere and in aquatic ecosystems. The response time to changes in atmospheric oxidized Hg input is most rapid, whereas the response to changes in Hg^0 and other environmental variables is much slower.

In terms of relative toxicity, MeHg is orders of magnitude more toxic than the inorganic forms of ionic Hg and Hg^0. Children of women who are exposed to MeHg during pregnancy are at risk of adverse neurodevelopmental outcomes. There is also some evidence of impacts of MeHg exposures on cardiovascular health, but this is less certain.

Fish consumption patterns differ across geographic regions and vary according to traditional diets, recreational activities, and proximity to supply of fresh fish. Individual variability in Hg exposures across populations reflects these differences as well as the types and origins of fish consumed. For some populations, most of the fish that are consumed are from local sources. In other populations, marine migratory fish, such as tuna or swordfish, or foreign-raised fish constitute the largest fraction of consumption. In regions that have not been contaminated by large local sources of Hg, the majority of population-wide human exposure to Hg is from marine fish consumption.

Hg concentrations in commonly consumed migratory marine fish, such as tuna and swordfish, are affected by intercontinental transport and deposition of Hg to marine ecosystems. Changes in open ocean Hg concentrations can be attributed in part to intercontinental atmospheric transport of Hg and localized deposition of Hg plumes from large source regions, such as the deposition of Hg emissions from Asia into the northern Pacific. In addition to atmospheric transport, large-scale oceanic transport can also be responsible for long-range transport of Hg from the original emissions source and likely impact marine fish concentrations globally.

Thus, to understand the main impact that emission controls will have on MeHg exposure over intercontinental scales, it is necessary to understand the linkages among atmospheric and oceanic transport, methylation in marine ecosystems, exposure and biomagnification in migratory marine fish, the capture and international trade of seafood, and seafood consumption patterns. These linkages are presently poorly quantified.

Adverse impacts on fish, birds, and mammals from recent and current anthropogenic Hg inputs into the environment have been documented. Efforts to monitor spatial and temporal trends of Hg in wildlife are underway and preliminary results suggest that intercontinental Hg transport contributes to adverse effects on ecological health.

Persistent Organic Pollutants

By definition, POPs are persistent, bioaccumulative, and toxic. Their adverse effects on human health and wildlife range from various forms of acute toxicity to chronic effects, including carcinogenicity and developmental and reproductive effects. It is the chronic effects from low dose exposures that are most relevant with respect to the impacts of intercontinental transport.

Similar to Hg, POPs are widely distributed through atmospheric transport, but their primary environmental impacts are realized through the contamination of food webs. Through processes of bioconcentration, bioaccumulation, and biomagnifications, humans and other animals may be exposed to much greater concentrations of POPs than observed in the air, water, or soil.

There is little information about long-term trends of POPs in food or human media outside of western Europe, North America, and Japan, making it difficult to characterize the global impacts of POPs on human health.

The traditional diets of indigenous Arctic people are high in locally caught fish and game. Combined with the atmospheric and oceanic transport of POPs into the Arctic and accumulation in the Arctic food web, such diets result in elevated POPs exposures in native Arctic populations. Studies have demonstrated elevated exposures to PCBs in native Arctic populations and have estimated that exposures to toxaphene, chlordane, DDT, HCH, dioxins and other dioxin-like POPs can be above levels of concern for adverse health effects. Moreover, there is concern about new POPs, which are currently in commerce and have yet to be addressed by international agreements, including penta-bromodiphenyl ethers (PBDEs), perfluorooctane sulfonate (PFOS), and perfluorooctanoic acid (PFOA), that have been detected in Arctic environments, in some cases in increasing amounts. The risks of chronic exposure to these chemicals are not well characterized.

In addition to elevated human exposures, studies have documented elevated concentrations of POPs in wildlife in remote environments. There is currently no strong evidence that Antarctic organisms or ecosystems are being impacted by organic contaminants associated solely with long-range atmospheric transport (and not associated with human activities at research bases). There are reasons to believe, however, that Antarctic organisms are more susceptible to the effects of organic contaminants than temperate organisms, though more research is clearly needed in this area.

5. How may the source-receptor relationships change over the next 20 to 40 years due to changes in emissions and climate change?

The significance of intercontinental transport for the achievement of environmental policy objectives may change in the future due to changes in the magnitude and spatial distribution of emissions. These changes may be caused by changes in technology and land use, the continuing implementation of pollution control measures, regional differences in the pace of economic development, the growth in shipping and aviation emissions, and the implementation of climate change mitigation measures. In addition, climate change may cause changes in transport patterns, precipitation, atmospheric chemistry, land cover, natural emissions, re-emission of legacy pollution, and anthropogenic primary emissions that affect the magnitude of intercontinental transport. The impact of expected changes on intercontinental transport of O_3, PM, Hg, and POPs is discussed below.

Given expected changes in emissions over the next 20 to 40 years, it is likely that it will become more difficult for some nations to meet their own environmental policy objectives without international cooperation to address transboundary and intercontinental flows of air pollution.

Ozone and Particulate Matter

The current levels of intercontinental transport and hemispheric baseline concentrations of O_3 and PM are a result of emissions that, on a global basis, increased rapidly between 1950 and 1990. Since 1990, global emissions leading to O_3 and PM concentrations have experienced little change or have begun to decrease. In Europe and North America, which have been the dominant sources of anthropogenic emissions until recent decades, emissions of most precursors are constant or declining, due to the implementation of air pollution control policies. In East Asia and South Asia, emissions of precursors have risen dramatically in recent years, due to economic growth and development in these regions.

The implications of changes in anthropogenic emissions were explored in the HTAP multi-model experiments by considering a set of global emission scenarios developed to inform the Intergovernmental Panel on Climate Change's (IPCC) fifth assessment report, known as the Representative Concentration Pathways (RCPs). Three of the four scenarios assume some climate change mitigation policy will be adopted, and all four assume that the implementation of air pollution control policies will increase as development and income increase. As a result, all four of the scenarios suggest that between now and 2050, global emissions of most O_3 and PM precursors will decline, up to 76% for SO_2 emissions. However, the regional distribution of emissions in the Northern Hemisphere is expected to shift, with steeper and earlier declines in Europe and North America and shallower declines or actual increases in South and East Asia. Under the lowest emissions scenario, NO_X emissions between 2000 and 2050 decline by 78%, 63%, and 48% in North America, Europe, and East Asia, respectively, but increase in South Asia by 42%. Under the highest emissions scenario, NO_X emissions peak in 2030 with decreases of 43% and 16% in North America and Europe, respectively, and increases of 65% and 91% in East Asia and South Asia, respectively.

As part of the HTAP multi-model experiments, the impact of this redistribution of future emissions and expected changes in future global CH_4 concentrations as specified by the RCP scenarios was explored using linear approximations of the intercontinental source-receptor sensitivities determined under current conditions. To illustrate the range of future levels of air pollution, we assessed the RAIRs for 2050 under the lowest emissions scenario and for 2030, when global emissions peak, under the highest emissions scenario. The results are compared to the 2001 base case in Table 5.1. For North America ground-level O_3 concentrations, the RAIR is estimated to increase to around 50% under both the high and low emissions scenarios, suggesting that, in the future, changes in emissions of O_3 precursors outside the region may be as important as changes within the region. For Europe, the RAIR for ground-level O_3 increases relatively little under the highest and lowest scenarios, due to the relatively similar emissions trajectories in Europe and North America. For East Asia, the RAIR decreases under a high emissions scenario, under which emissions within the region increase, and increases under a low emissions scenario, under which emissions in the region decline. For South Asia, the RAIR for ground-level O_3 declines under both the low and

high scenarios. Note that the sensitivity to changes in geographic distribution of emissions within a particular region has not been adequately assessed yet.

Table 5.1. The Relative Annual Intercontinental Response (RAIR) of pollutants to 20% emission decreases in four regions approximating North America, Europe, South Asia, and East Asia, under three scenarios: a 2001 base case, a high global emissions scenario in 2030, and a low global emissions scenario in 2050.

Scenario	Pollutant/Parameter[a]	Receptor Region			
		North America	Europe	South Asia	East Asia
2001 base[b]	O_3 Concentration	32%	42%	33%	40%
2030 high[c]	O_3 Concentration	53%	45%	20%	30%
2050 low[d]	O_3 Concentration	49%	44%	14%	41%

Notes:
[a]   Concentrations are at ground-level.
[b]   Calculated from linear approximations of a subset of models participating in the HTAP multi-model experiments.
[c]   Based on Representative Concentration Pathway 8.5, a high emissions scenario.
[d]   Based on Representative Concentration Pathway 2.6, a low emissions scenario

Using the linear approximation, the influence of changing CH_4 concentrations can be separated from the influence of changing intercontinental transport and local and regional emissions over the historical emission trends and future emission scenarios, as shown in Figure 5.1. Roughly 40% of the O_3 increase since the preindustrial period is believed to be due to anthropogenic CH_4. By 2050, the RCP scenarios suggest a wide range of possible changes in CH_4 concentrations, ranging from an increase greater than 50% to a decrease greater than 15%. Under the highest scenario, expected increases in CH_4 concentrations have a large influence on ground-level O_3 changes, in some cases offsetting significant decreases in O_3 formation associated with local and regional emissions.

The HTAP multi-model experiments also examined the potential impacts of changes in meteorology and transport patterns expected as a result of climate change on O_3 concentrations using a set of three models that simulated climate changes between the periods 2000 and 2100. Future changes in climate are expected to increase the effect of O_3 precursor emissions over source regions and reduce the effect over downwind receptor regions. However, the magnitude of these effects is relatively small, and is driven mostly by changes in atmospheric chemistry and not by changes in transport patterns. The effect of natural emission changes and wider climate-related feedbacks are potentially important, but have not been evaluated fully yet.

Mercury

Two recent studies have developed global emission projections for anthropogenic Hg emissions in the years 2020 and 2050. One of the studies was based on the IPCC Special Report on Emissions Scenarios (SRES), used in previous IPCC assessments. The other study was developed for the United Nations Environment Programme's (UNEP's) Mercury Programme. Both studies conclude that significant increases, up to 25% in 2020 and 100% in 2050 as compared to 2005, in global Hg emissions can be expected if no major changes in emission controls are introduced and economic activity continues to increase. In both studies, the largest increase in emissions is projected from coal combustion for electricity generation in Asia. However, the implementation of available emission control technology could stabilize or decrease these emissions.

a) Mean ground-level O₃ changes under 4 scenarios
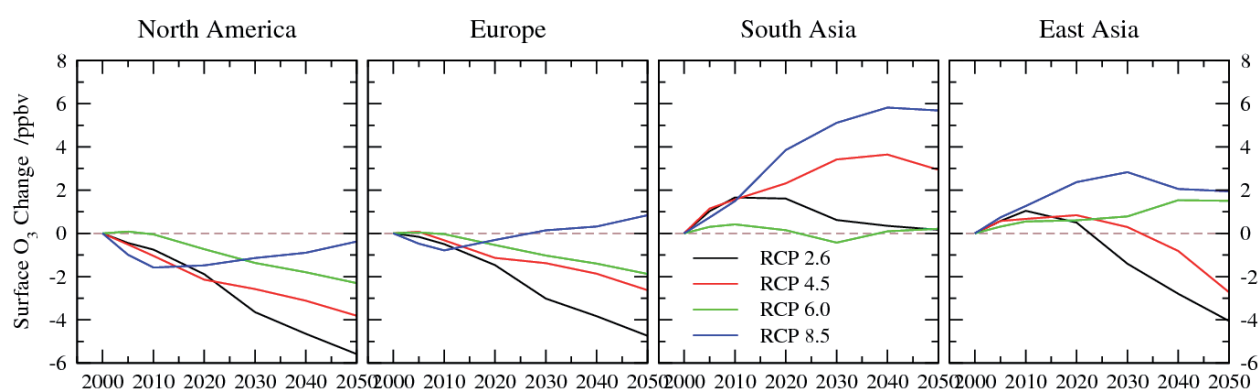
b) Contribution to mean ground-level O₃ changes of regional emissions, intercontinental transport, and CH₄.
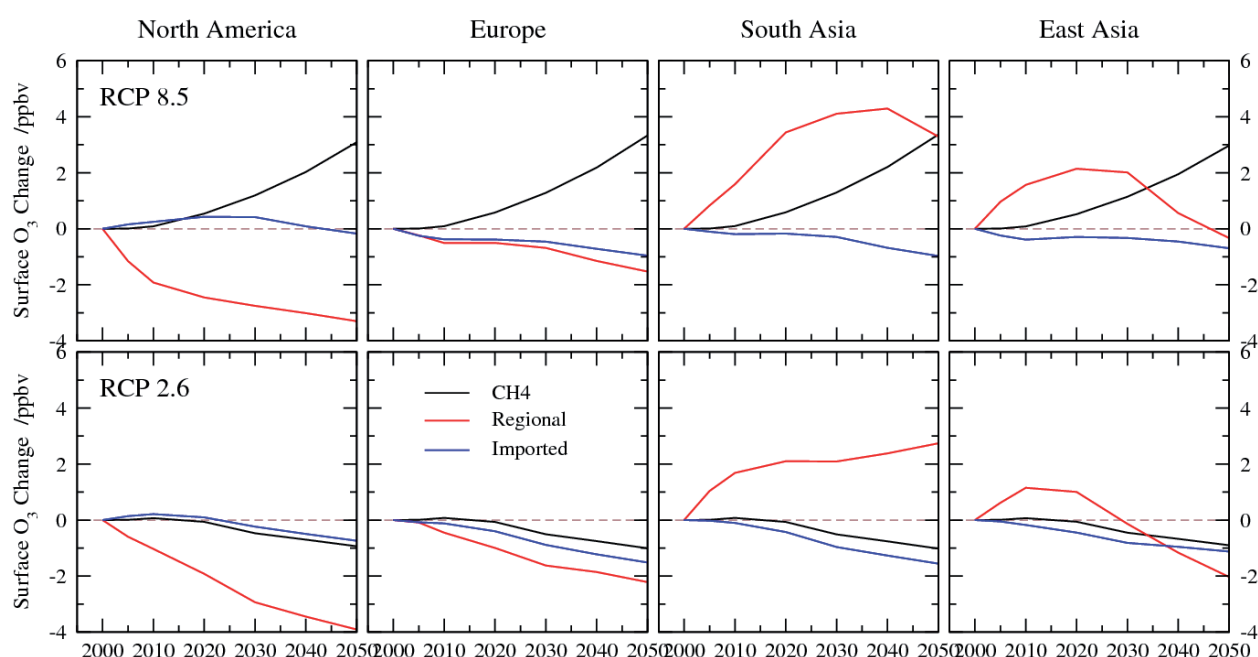
Figure 5.1. The role of intercontinental transport and CH4 in future O3 scenarios. a) Mean surface O_3 changes over the four HTAP receptor regions following the four RCP scenarios from 2000 to 2050, based on linearization of the six models that contributed results to all SR simulations in the HTAP multi-model experiments. b) Contribution to annual mean surface O_3 changes for RCP 8.5 (top row) and RCP 2.6 (bottom row) scenarios from changes in CH_4 concentration, emission changes over the receptor region ('Regional') and emission changes from outside the receptor region ('Imported').

As part of the HTAP multi-model experiments, three of the scenarios for 2020 were simulated by the participating models. The scenarios ranged from a 25% increase to a 65% decrease in global emissions over 2005 levels. The participating models predicted corresponding increases in deposition of 2-25% and decreases in deposition of 25-35% in four source regions, respectively, in annual simulations. In remote regions, such as the Arctic, the changes were predicted to be smaller – ranging from 1.5-5% increases to 15-20% decreases.

The intercontinental source-receptor relationships under these future scenarios are not significantly different from the source-receptor relationships estimated for current emissions. The large contribution of natural sources and re-emitted legacy Hg to deposition dampens the relative response of Hg deposition to changes in new anthropogenic emissions, reinforcing the long term benefit of decreasing the amount of Hg re-circulating in the environment by decreasing Hg emissions globally now.

The impacts of climate change on intercontinental transport of Hg were not explicitly addressed in the HTAP multi-model experiments, and there is large uncertainty about how climate change will effect natural and recycled emissions of Hg, as well as the atmospheric chemistry and transport of Hg. Climate change effects on temperatures, frequency of forest fires, plant growth and decomposition will significantly impact the terrestrial-atmospheric exchange of Hg. Likewise, ocean-atmosphere exchange of Hg will be affected by changes in temperature, wind speeds, storm frequency, as well as changes in atmospheric oxidant and aerosol concentrations. The net effect of these changes, however, has not been adequately studied yet.

Persistent Organic Pollutants

Over the next 40 years, emissions and patterns of intercontinental transport of some POPs will continue to decrease and shift as a result of national and international regulations. For those POPs whose use has been banned or strictly limited, geographic shifts in transport may occur as re-emission of legacy pollution results in migration or dispersion of the pollutants. For POPs that are still in use as chemicals or are unintentionally released from combustion or other industrial processes, differences in regulations or economic activity may lead to shifts in the spatial distribution of emissions. In Europe and North America, full implementation of the LRTAP POPs Protocol, the Stockholm Convention, and other national legislation is expected to decrease emissions by more than 90% for hexachlorobenzene (HCB) and PCB, more than 60% for pentachlorodibenzodioxins and furans (PCDD/Fs), and 30-50% for PAHs.

For POPs that are still in commerce and have yet to be addressed by national and international regulations, intercontinental flows are expected to increase as continued emissions contribute to the stock of the pollutant circulating in the environment.

Climate change may further alter the magnitude and patterns of emissions and intercontinental transport of POPs. Similar to Hg, climate change may significantly alter the exchange of POPs among the atmosphere and water, soil, vegetation, sediments, snow, and ice. There is evidence that climate change phenomena, e.g., elevated temperatures and sea-ice reduction, and extreme climate change induced events, such as forest fires, flooding and glacial melting, will remobilize POPs previously deposited in sinks, e.g., forest soils and vegetation, ocean and lake sediments and glaciers. Climate change may also alter the exposures of individuals and populations and their vulnerability to chemical exposures.

6. How well can we represent the processes that affect these intercontinental or global flows of air pollutants in quantitative models?

Our ability to represent the processes that drive intercontinental or global flows of air pollutants in quantitative, predictive models varies across the different pollutants of interest. In general, our confidence in the predictions of the models decreases from O_3 to PM to Hg to POPs, as the complexity of the processes that must be represented increases and as the available observational data base decreases.

We can evaluate our modelling capabilities by comparing model estimates of concentrations and deposition to the observed magnitudes, patterns, and trends for each pollutant. We can also compare the estimates of different models, which give us some sense of the lower bound of the uncertainty in our modelled estimates. Estimating the current level of uncertainty or confidence in modelled estimates of intercontinental source-receptor relationships and identifying major areas of uncertainty have been main objectives of the HTAP multi-model experiments.

Ozone

Model to Observation Comparisons

Current global O_3 models reproduce much of the observed regional and seasonal variability in surface concentrations and deposition and have been used to predict the path of pollutants during individual intercontinental transport events. This ability gives us some confidence that we can quantitatively represent the key processes controlling the formation, transport and removal of O_3 and its precursors. However, on finer spatial and temporal scales, significant discrepancies exist among estimates from different models and between model estimates and observations, indicating weaknesses in our representation of many processes at the resolution of current models. Furthermore, current global models are not able to reproduce entirely historical O_3 measurements and observed trends, raising concern about our ability to predict changes in the future.

As part of the HTAP multi-model experiments, the ensemble of O_3 simulations was compared to O_3 concentrations observed by selected surface networks in North America, Europe, and Asia. At most sites, the model ensemble mean generally captures the observed seasonal O_3 cycle and is close to the observed regional mean surface O_3 concentration. Furthermore, the model ensemble mean agrees well with regionally averaged observed values in all regions in spring and late autumn when intercontinental transport tends to be strongest.

However, there are notable biases in the model ensemble mean. At high altitude sites, which are more likely to be sampling the free troposphere and are better suited for detecting intercontinental transport, the ensemble mean tends to underestimate observed surface O_3 concentrations. Furthermore, the multi-model mean overestimates the observed summertime surface O_3 concentrations over Japan (with a bias of 12 ppbv) and the eastern United States (with a bias greater than 15 ppbv).

The ability of the models to estimate maximum daily 8-hour average O_3 concentrations, was evaluated by comparing the model estimates to observed concentrations at Clean Air Status and Trends Network (CASTNet) sites in the United States. The multi-model ensemble mean is able to capture the overall pattern of observed daily maximum 8-hour average O_3 concentrations observed at rural sites in the United States with a bias of +4.1 ppbv considering all seasons. However, large positive mean biases exist during summer for the eastern United States, ranging from 10-20 ppbv.

In addition to comparison to surface observations, the results of the HTAP multi-model experiments were also compared to observations from sondes (or balloons). The ability of current models to reproduce O_3 measurements aloft is not as good as it is at the surface, but the ensemble mean is able to reproduce much of the observed seasonal cycle. The seasonal averages for most models are within 20% of the sonde measurements for O_3 concentrations in the lower and middle troposphere. For the upper troposphere, the bias is often higher. Model performance varies by location and season. For some polar sites, there are virtually no correlations between models and O_3 sonde measurements in winter and spring at all altitude levels, and the range of the model estimates is large.

As with surface concentrations, the models do not capture well the observed day-to-day variability of O_3 concentrations aloft.

Model to Model Comparisons

In the HTAP multi-model experiments, the differences among models in the response of O_3 concentrations due to 20% changes in precursor emissions are appreciable (with a standard deviation of 20% to 50% of the ensemble mean response) in both the source region and at distant receptors. Across the northern mid-latitudes, the differences among models are largest for changes in European emissions, with a standard deviation of about 0.2 ppbv, which is the same order of magnitude as the mid-latitude annual mean response. The standard deviation of the model responses is also about 0.2 ppbv over the North Atlantic and in Western Europe for changes in North American emissions and over the western North Pacific for changes in Asian emissions, but for these cases, the standard deviation is generally less than half of the multi-model annual mean decrease in surface O_3.

The range of the estimated source-receptor relationships across the models participating in the HTAP multi-model experiments is generally smaller than the range of estimates found in previous studies in the literature, but the HTAP multi-model experiments estimates may fall within, bracket, or fall outside the range from previous studies. The HTAP multi-model experiments estimates are generally lower than the highest estimates of intercontinental transport found elsewhere in the literature. These higher values were estimated from simulations in which anthropogenic emissions were set to zero, or in which any O_3 production throughout the tropospheric column over a source region was considered to represent the effect of emissions from the source region. The estimates from the HTAP multi-model experiments are lower as they are based on scaling up 20% changes in anthropogenic emissions from four main source regions.

Sources of Uncertainty

The differences across models and between models and observations are due to uncertainties in emissions estimates, limitations imposed by the spatial and temporal resolution of the models, and uncertainties in the representation of transport, chemistry, and removal processes. Each of these sources of uncertainty is discussed briefly below.

The quality of emissions inventories varies widely, by pollutant, by region, and by source category. Estimates of uncertainties in anthropogenic NO_X emissions range from ±14% for Europe to ±37% for Asia. Uncertainties for VOCs can be much higher. Significant uncertainties in NO_x, VOC, CH_4, and CO emissions inventories are associated with inadequate knowledge of natural emissions (including various CH_4 sources and soil and lightning NO_x), open biomass burning (forest and grassland fires, and agriculture waste burning), biofuel use (for heating and cooking), artisanal industry, residential combustion of coal, and agricultural production systems.

The models participating in the HTAP multi-model experiments each used their own best estimate of annual emissions. The total global emissions for some O_3 precursors vary significantly across the different models, with ranges as high as 50% of the ensemble mean values.

Current global models are based on grids with cells larger than 100 kilometres in the horizontal dimension. While large-scale transport patterns are reasonably well represented by these models, processes that occur on finer spatial scales are not well parameterized. These processes include frontal lifting, deep convection, atmospheric boundary layer mixing, atmospheric boundary layer venting, and wet deposition. These processes drive the continental export of pollution and can create well-defined, observable plumes that maintain their identity for a week or more. At the resolution of current models, the distinct nature of these plumes is lost too quickly. Thus, current models are unable to reproduce the fine structure and variability in transport observed in the atmosphere.

The processes of stratosphere-troposphere exchange and subsidence of pollution aloft back into the boundary layer are also not well-characterized, affecting the ability of current models to estimate concentrations aloft and the impact of transport at the surface.

Global models are not well suited to estimate concentration responses at urban scales, as the resolution of current models is too coarse to resolve the strong chemical contrasts associated with urban regions. These limitations may be addressed in the future by coupling global models with higher-resolution regional models or by using novel methods such as adaptive grids or embedded plume models.

Significant uncertainties also exist in the photochemical mechanisms used in current models, including the treatment of dinitrogen pentoxide (N_2O_5) hydrolysis, peroxyacetyl nitrate (PAN) formation and sinks, isoprene chemistry, and nitrate (NO_3) nighttime chemistry. The uncertainty in chemical mechanisms employed in models is equivalent to the uncertainty induced from the missing resolution-dependent processes in the global model and have been shown to lead to large differences in regional model results.

Lastly, removal processes of wet scavenging and wet and dry deposition are not well parameterized at the scale of global models.

In the HTAP multi-model experiments, a series of standardized process tracer studies (TP) were conducted to begin to understand differences in the model results that can be attributed to the driving meteorology or parameterization of transport processes. Preliminary results from these experiments suggest that there are significant differences among models in the timescales for transport of pollutants away from surface sources.

Conceptually, the various sources of uncertainty can be classified based on 1) the magnitude of uncertainty and 2) the likelihood that the uncertainty will affect estimates of source-receptor relationships or source attribution for O_3 and PM on intercontinental scales. Figure 6.1 presents one attempt based on expert judgment to classify the sources of uncertainty in this two dimensional space. The top right quadrant of the figure indicates those aspects that are important, but are better quantified. The top-left quadrant indicates major elements contributing to the uncertainty in current model estimates of source-receptor relationships and source apportionment and identifies where research is needed to improve (reduce uncertainty) estimates of intercontinental transport.

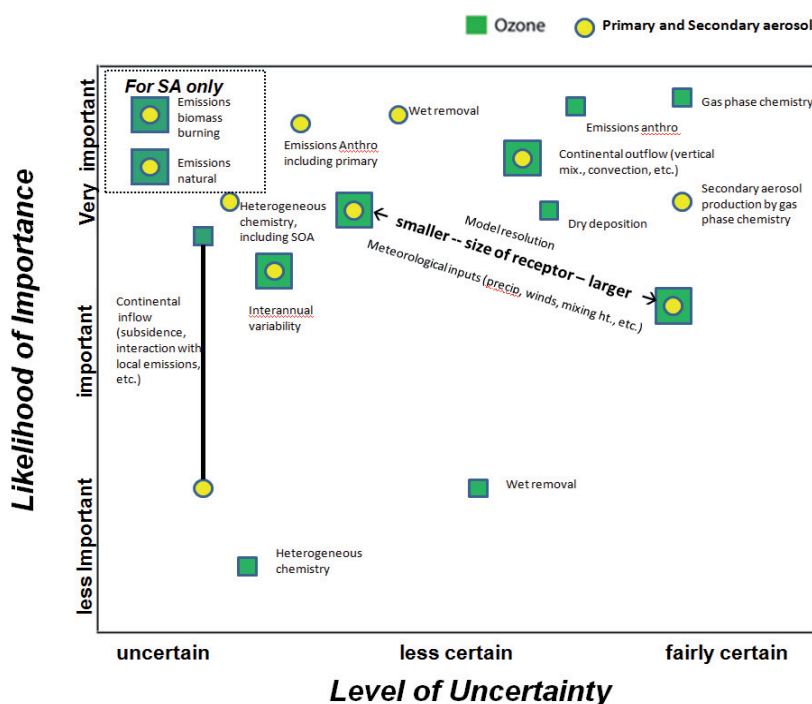**Intercontinental Source Attribution (SA) and Source-Receptor (S/R) relationships**

Figure 6.1. Classification of sources of uncertainty in global model estimates intercontinental source-receptor relationships and source attribution (SA) for O_3 and PM based on expert judgment.

Particulate Matter

Many of the same limitations and sources of uncertainty faced in estimating O_3 concentrations apply to current models for PM, although there are additional challenges as well. In the HTAP multi-model experiments, the range of predicted concentrations across the participating models varies across the chemical components of PM. The differences across models are about a factor of two for sulphate surface concentrations in the mid-latitude continental regions, whereas the model differences approach a factor of four for black carbon and POM surface concentrations, and a factor of seven for soil dust surface concentrations. For the Arctic region, where PM levels are very low, the relative spread of modelled concentration estimates is much greater. A detailed comparison across the HTAP-participating models reveals a factor of four difference in the atmospheric lifetime of sulphate calculated by the different models.

A comprehensive comparison of model estimates to observations has not been conducted. However, available observational data to support such an evaluation are being compiled by the Norwegian Institute for Air Research (NILU) (which hosts the European Monitoring and Evaluation Programme-Chemical Coordinating Centre or EMEP-CCC under the LRTAP Convention) as part of the EBAS-HTAP observations database and some comparisons have been conducted for sulphate wet deposition.

For PM, the same limitations of coarse model resolution apply, making it difficult for current models to reproduce the fine structure and variability of transport observed in the atmosphere.

The uncertainty in the emissions inventories used in the models varies widely, by chemical component, by region, and by source category. Estimates of uncertainties for anthropogenic emissions range from ±5% for SO_2 emissions in Europe to a factor of four for black carbon emissions in Asia. Major uncertainties in emission inventories are associated with inadequate knowledge of natural emissions (including volcanoes and windblown mineral dust), open biomass burning (forest and grassland fires, and agriculture waste burning), biofuel use (for heating and cooking), artisanal industry, and residential combustion of coal. These propagate into higher uncertainties in emissions for the pollutants that are mainly associated with these activities, such as soil dust, POM, and black carbon.

The models participating in the HTAP multi-model experiments each used their own best estimate of annual emissions. The emission estimates for individual PM components from specific regions can vary across models by more than a factor of two.

In addition to the uncertainties associated with emissions and transport processes, significant uncertainties in the model estimates are associated with secondary organic aerosol formation and dry and wet deposition processes.

Mercury

Current global atmospheric Hg models reproduce the observed ground-level Hg^0 concentrations to within 20% of the sparse observations that are available and reproduce the pronounced inter-hemispheric gradient in baseline Hg^0 concentrations that has been observed. The agreement between models and observations for Hg wet deposition is weaker, with differences between observed and modelled values up to 100%, mainly due to uncertainties in Hg emission rates, Hg oxidation chemistry and estimated precipitation rates. There are larger differences across model estimates of wet deposition in areas where there are little observational data. Significant differences exist among models for dry deposition, which is believed to contribute as much as wet deposition to total Hg deposition and for which there is little observational data.

Using identical inputs for new anthropogenic emissions, the four models participating in the HTAP multi-model experiments predict comparable Hg deposition levels in Europe, North America and South Asia, but the estimated Hg deposition levels are a factor of 1.5 higher in East Asia. The level of agreement is striking, given the significant differences among models in the assumptions used concerning emission rates for natural or re-emitted Hg and the oxidation pathways for Hg^0. For

deposition in the Arctic, the differences across models are much higher, with a spread greater than a factor of four, due to differences primarily in the representation of atmospheric Hg depletion events.

The relative contributions of the major source regions to region-wide average Hg deposition in different regions are very similar among the models. The most significant deviations in the modelling results are seen in areas with large anthropogenic and natural and secondary emissions. This is due to large uncertainties in the natural and secondary emission estimates and to the differences in spatial resolution of the participating models, which varied by a factor of four.

Although there are significant uncertainties in the anthropogenic emissions from some source categories and regions of the world, the magnitude of new anthropogenic emissions is thought to be much less than the emissions of re-emitted Hg that was previously deposited. However, the rates of re-emission from terrestrial and aquatic systems, especially from the ocean which may emit twice as much Hg as anthropogenic sources, are not well characterized.

The magnitude of annual Hg emissions is relatively small compared to the reservoir of Hg^0 already in the atmosphere. The reaction pathways by which this Hg^0 is oxidized are not well understood. It is not clear what fraction of oxidation is due to reaction with O_3, hydroxyl radical, bromine containing compounds, or other oxidants. There is little consensus on the rates, temperature dependence, and products of the reactions of Hg with these oxidants, and how their relative magnitudes differ in different environmental conditions, such as the polar spring, marine boundary layer, or upper troposphere. Similarly, there is little consensus about the pathways for the chemical reduction of oxidized Hg in the atmosphere.

The health and environmental risk associated with Hg is primarily associated with exposure through contaminated fish. Thus, to draw the connection between source and receptor it is necessary to not only simulate the atmospheric fate of Hg, but also to simulate its deposition; its methylation in aquatic ecosystems, including the ocean; its bioconcentration, biomagnifications, and bioaccumulation in aquatic food webs; and exposure through internationally traded fish. There are significant uncertainties in our ability to simulate each of these steps, as well.

Persistent Organic Pollutants

As with Hg, POPs models must not only simulate the behaviour of pollutants in the atmosphere, but they must also simulate the exchange between the atmosphere and other environmental media (such as water, soil, snow, ice, and vegetation) and the transport and transformations that occur in those other media. Observational data from these media are limited, making it difficult to evaluate models and characterize uncertainties.

Current POPs models vary widely in the level of detail represented. Model simulations for a subset POPs have been conducted and are typically able to reproduce observed annual concentrations to within a factor of three or four, enabling identification of major transport pathways. The POPs that have been successfully modelled and evaluated include selected PCBs, HCHs, and PAHs. In some cases, however, the differences between model estimates and observed values can be much greater indicating fundamental uncertainties both in emission inventories and in modelling approaches.

For some POPs, there are significant uncertainties associated with the pollutant's physical-chemical properties, such as Henry's Law constants, vapour pressures, and octanol-air partition coefficients. These physical-chemical parameters are used in the models to predict how the pollutant will move among media. However, the total environmental lifetime and long-range transport potential of a POP are not intrinsic substance properties. The fate of pollutants also depends on the characteristics of the environment in which it is found. Little is known about how the abundances and biodegradation of POPs vary in heterogeneous media such as soil, snow, oceans, and lakes.

7. What efforts are needed to develop a system of observations, emissions, and models to better understand and track these flows?

Our current understanding of the magnitude of intercontinental flows of air pollution is sufficient to conclude that such flows have a significant impact on environmental quality throughout the Northern Hemisphere and that coordinated international actions to mitigate these flows would yield significant environmental and public health benefits. However, our current ability to accurately and precisely quantify the contribution of intercontinental flows on air pollution concentrations or deposition at any given location or the effect of international emission controls on pollution levels and their environmental or public health impacts is limited. Important areas of uncertainty are identified in the answer to Question 6.

To be able to better quantify the impacts of intercontinental flows of air pollution and the effects of international controls, additional efforts are needed to improve the coverage and resolution of our observational systems, the accuracy and resolution of our emissions inventories and projections, the fidelity and performance of our models of chemistry and transport, and the scope and detail of our impact assessments. Moreover, we need to shift the goal of the science activity from simply developing knowledge to a goal of informing action. Specific recommendations for future work are provided by pollutant and discipline (i.e., observations, emissions, atmospheric modelling, and impact assessment) in the chapters of Parts A, B, and C of this assessment. Taken together, these recommendations present a description of a programme of monitoring, research, and analysis activities that must be intentional, innovative, integrated, supported by institutions and information networks, and inclusive. These concepts are discussed in more detail below.

By intentional, we mean that the goal of quantitatively attributing concentrations, deposition, and impacts to emission sources and estimating the effect of emission changes on intercontinental scales must be an explicit part of the design objectives for future efforts. Characterizing continental pollution inflow and outflow have been stated design objectives for a number of intensive field campaigns and the operation of a few remote monitoring sites. However, most existing observational systems have been designed to characterize the spatial and temporal distribution of pollutants and track pollution trends over time. To improve our ability to understand intercontinental transport, observational systems need to be designed to collect additional information needed to quantitatively attribute pollutants to their sources. Intensive field campaigns and routine monitoring programmes need to be designed to collect information that can be used to evaluate the representation of emission sources, transport and chemical processes, and air-surface exchanges in current models.

By innovative, we mean that future efforts must employ novel techniques and develop new methods where needed. Observational programmes and intensive campaigns must take advantage of new fingerprinting techniques that rely on detailed chemical speciation or isotopic or chiral signatures. Observations from satellites need to be more fully exploited to understand transport patterns, trends, and events and provide a basis for evaluating models and emission inventories. New techniques need to be developed to quantify important processes, such as dry deposition, air-surface exchanges, or chemical speciation (e.g., speciation of Hg oxidation products). New modelling techniques such as adjoint analyses, two-way nesting, and plume-in-grid algorithms need to be employed to better represent transport processes and understand source attribution and source-receptor relationships.

By integrated, we mean that future efforts must generate new insights by combining and comparing information within disciplines and across disciplines, as well as across different pollutants (i.e., O_3, PM, Hg, POPs). The elements of an integrated effort are depicted in Figure 7.1. Within the field of emissions estimation, emission inventories developed at different spatial scales or with different methodologies must be compared and reconciled. Emissions for different pollutants should be compared for consistency where the underlying emission generating activity is the same. Through the use of inverse modelling, emissions must be estimated from observations and compared to activity-based estimates. Within the field of observations, data from different networks, analysis methods, or platforms or data for different pollutants must be brought together to create richer descriptions of the state of the atmosphere and how it is changing. Such richer data sets must be used

to evaluate models of transport and chemistry or improve model predictions through data assimilation techniques. Multiple models of the same type, pollutants, and spatial scales, as well as models of different types, pollutants, and spatial scales need to be compared to understand the effect of representing different processes, employing different algorithms, and other modelling choices.
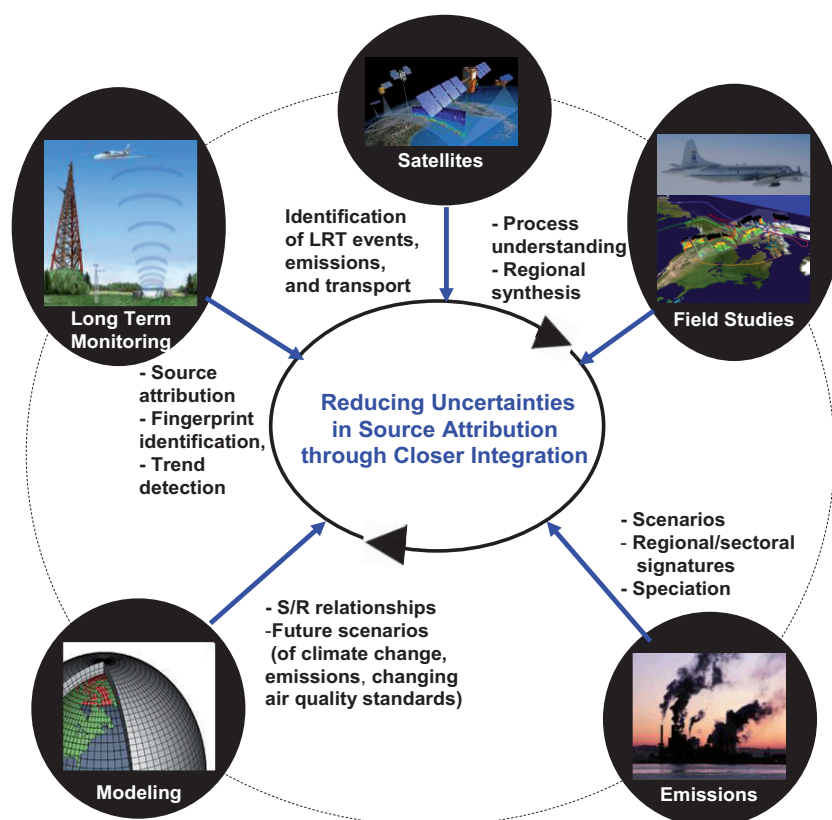
Figure 7.1. Major components of an integrated approach for source attribution related to long range transport (LRT). The circles denote the major components, the accompanying text indicates the major source and attribution-related outputs. Model and measurement elements refer to both meteorological and chemical components. [Reprinted with permission from Figure 6.1 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., USA.]

The different disciplines, pollutants, and spatial scales are often addressed by different expert communities. The integrating activities described above require institutional mechanisms to foster cooperation among individuals within these distinct communities, as well as among individuals in different communities. These mechanisms include international cooperative programmes, funded projects that require specific types of cooperation, or forums such as workshops that try to stimulate and facilitate cooperation. Many institutions already exist that address integration of activities across one or several relevant dimensions. These existing institutions include:

- global cooperative programmes such as the World Meteorological Organization's (WMO's) Global Atmospheric Watch (GAW) programme and Integrated Global Atmospheric Chemistry Observations (IGACO) Strategy, the Stockholm Convention's Global Monitoring Plan, the UNEP Mercury Programme, the International Geosphere-Biosphere Programme's (IGBP's) International Global Atmospheric Chemistry (IGAC) activity and the Global Atmospheric Pollution Forum

- regional cooperative programmes, such as the LRTAP Convention, East Asia Network for Acid Deposition, Malé Declaration, the Arctic Council, and others.

To be efficient, efforts to better understand intercontinental flows of air pollution must build upon these existing institutions, creating linkages among them where necessary.

The TF HTAP has played a constructive role in bringing together often separate expert communities across disciplines, pollutants, and spatial scales, and making connections among existing cooperative programmes. Sometimes the connection is made by simply co-organizing a joint meeting and identifying some follow-on collaborative activities. Sometimes the connection requires a more concerted effort to jointly plan activities. In the context of intercontinental transport, cooperation is only just beginning across a number of the communities that intersect as part of the TF HTAP. Continued effort is needed to bring experts from different communities together and more financial support is needed for their collaborative work.

In addition to institutional support, the integration of information across disciplines, pollutants, and scales needs to be supported and facilitated by improvements in the way communities of experts share and manage information. The Group on Earth Observations (GEO) is working to develop a Global Earth Observations System of Systems (GEOSS) that uses advanced information technology to lower the technological barriers to data access, exchange, integration, and analysis for a wide variety of earth science information. The assessment of intercontinental flows of air pollutants is an excellent test case for the development of such an information system of systems. The TF HTAP has been contributing to the development of GEOSS through the development of nomenclature conventions for atmospheric chemistry modelling, data repositories for modelling and observational data, and the development of standardized protocols for automated unattended access to these repositories. By continuing to invest in an information management infrastructure, we can improve the information base for future assessments, increase the efficiency of collaboration, and broaden the community that is able to participate in the assessment.

The community of experts that currently is engaged in understanding the intercontinental flows of air pollution through the TF HTAP and other institutions is relatively small, consisting primarily of global atmospheric science researchers, a majority of whom are from Europe and North America. To be successful in moving the issue beyond the realm of science and into the realm of policy action, a much larger and more diverse community must be engaged in understanding the issues and in considering management alternatives. This broader community must include scientists from relevant disciplines and air quality management officials in developed and developing countries throughout the Northern Hemisphere. Improving access to information through the development of GEOSS and building linkages among existing multi-lateral institutions in which scientists and air quality officials participate can help broaden the participation in future assessments. By creating an actively inclusive process for assessing intercontinental transport of air pollution, we can improve the likelihood that the findings of future assessments lead to international action to address the sources of transported air pollution.

8. What are the potential benefits of further international cooperation to address intercontinental transport of air pollution and how might this cooperation be structured?

O$_3$, PM, Hg, and POPs are significant environmental problems in many regions of the world. Mitigation of intercontinental transport is not a substitute for emission reductions at the local and regional scale. In most cases, concentrations within a source region are more sensitive to emission changes within that region. However, without further international cooperation to mitigate intercontinental flows of air pollution, it is likely that many nations will not be able to meet their own goals and objectives for protecting public health and environmental quality over the next 20 to 40 years.

Intercontinental flows of O$_3$ and its precursors have led to increasing trends in baseline O$_3$ concentrations measured at a number of remote sites across the Northern Hemisphere. The O$_3$ concentrations in the air masses moving from one region to another may approach or exceed current air quality standards or objectives in the downwind region. For example, the inflow to Europe from the North Atlantic can exceed the threshold of the LRTAP Convention's objective for vegetation protection. For PM, concentrations in the air masses carried by intercontinental transport may exceed short-term standards for human health protection in Europe and Asia and interferes with the attainment of long-term goals for visibility protection in western North America. For Hg, coordinated action is needed to decrease the global pool of atmospheric Hg and the amount of Hg cycling through the environment. For POPs, intercontinental flows introduce toxic contaminants from one region of the world to the environment and food web in another where local sources do not exist or have been eliminated.

Efforts to mitigate the sources of intercontinental flows of air pollutants result in significant benefits for both the source and receptor countries in terms of decreased impacts on public health, decreased damage to ecosystems, and depending on the mix of pollutants, decreased contribution to climate change. In fact, countries outside of a source region may benefit collectively more from emission decreases in a source region than the source region itself. One country's actions can also lessen the costs of emissions control needed in other countries. Thus, there are significant benefits to both source and receptor countries to cooperate in decreasing emissions that contribute to intercontinental transport of air pollution.

As discussed previously in response to Question 7, continued research, monitoring, and analysis efforts that are intentional, innovative, integrated, and inclusive are needed to further improve our collective understanding and quantification of the intercontinental transport of air pollution and its impacts. As we move from developing knowledge to informing collective action, the costs and availability of controls for emission sources and how they differ across and among continental source regions also need to be assessed further. Many control strategies effect the emissions of multiple pollutants simultaneously; for example, strategies that decrease coal combustion will affect emissions of O$_3$ and PM precursors, as well as emissions of Hg, dioxins, and furans, each having multiple impacts over multiple spatial scales. In future work, the implications of different levels of control on different source sectors in different locations need to be examined holistically in terms of their multiple impacts on public health, ecosystems, and climate change at the local, regional, and global scales.

The availability of forums for pursuing further international cooperation to mitigate sources of intercontinental transport differs depending on the pollutants of interest. For POPs, the need to cooperate globally has been recognized by the 172 nations that are Parties to the 2001 Stockholm Convention on POPs. For Hg, the need to cooperate further was acknowledged in 2009 by 141 countries of the UNEP Governing Council in launching negotiations on a legally-binding instrument to address global Hg pollution. Thus, the Stockholm Convention and the ongoing global Hg negotiations provide forums for furthering global cooperation to mitigate sources of POPs and Hg.

For O$_3$ and PM, however, there is not yet an international agreement addressing global or intercontinental flows of pollution across the Northern Hemisphere. Instead, there are a series of regional agreements, in varying states of evolution, that address at least some of the sources of transported O$_3$ and PM. The 1979 LRTAP Convention is the largest and most mature of these regional agreements, with 51 Parties covering the regions of North America, Europe, and Central Asia. The Acid

Deposition Monitoring Network in East Asia (EANET), comprised of 12 Parties, and the Malé Declaration on Control and Prevention of Air Pollution and its Likely Transboundary Effects for South Asia, comprised of 8 Parties, were initiated formally in 1998. The UNEP Regional Resource Center for Asia and Pacific has been instrumental in establishing some coordination among EANET, the Malé Declaration, and other air pollution efforts in the region. Through the activities of the Global Air Pollution Forum, the creation of regional cooperative programmes related to O_3, PM, and their precursors have been explored recently in other parts of the world, including Africa and Latin America.

A range of approaches has been suggested for establishing global or hemispheric scale cooperation on mitigation of O_3 and PM, including negotiating a new international agreement; incorporating O_3 and PM into an existing global agreement, such as the United Nations Framework Convention on Climate Change (UNFCCC) or the Vienna Convention on the Protection of the Ozone Layer; expanding the geographic scope of the LRTAP Convention; or developing a global framework for cooperation within existing regional agreements. Although each of these approaches has advantages and disadvantages, in the near-term, the last approach is arguably the most promising for building the understanding and capacity needed to address air pollution problems on the local, regional, and global scales.

Existing regional cooperative efforts have already created organizational infrastructures and intergovernmental relationships upon which a global confederation, or inter-regional partnership, could be built to address air pollution issues at the local, regional, and global scales, building on the informal network established under the Global Air Pollution Forum. The regional efforts could benefit from two-way exchanges of information between the regions. Regions with more mature technical capabilities could provide other developing regions with assistance on emissions estimation, monitoring methodology, modelling techniques, impact assessment, strategy evaluation, management and control programme implementation, and enforcement. Developing regions, in return, could provide better information about the sources, character, and flow of pollutants originating in or affecting their regions. A global confederation could facilitate the connection among the regional agreements and global agreements and institutions, such as the UNFCCC, Vienna Convention, Stockholm Convention, and a potential global Hg convention, as well as UNEP, WMO, International Maritime Organization (IMO), International Civil Aviation Organization (ICAO), and GEO. Further, a global confederation could also facilitate the connection between the national and regional air quality management communities, represented in the regional programmes, and the global scientific community, as represented through such organizations as the IPCC and IGBP. Thus, a global confederation of regional cooperative programmes could help develop a better and globally-shared understanding of air pollution problems and their solutions at the local, regional, and global scale while maintaining autonomy and flexibility for regions to develop policies and programmes appropriate for their circumstances.

Given the work of the Global Air Pollution Forum, the TF HTAP, and UNEP, much of the foundation for a global federation or inter-regional partnership to address air pollution has already been laid. As a subsidiary body of the LRTAP Convention, the TF HTAP can continue to play a leadership role in bringing together different expert communities and connecting different regional and global institutions. For the LRTAP Convention, continuing and expanding such efforts is important to decrease the impact of sources outside the Convention and to meet the Convention's own goals. However, the potential for a more formal global federation of regional cooperative programmes on air pollution should be explored.

Whether formally or informally, science cooperation at the intercontinental scale must be expanded and improved to further our collective understanding of the intercontinental transport of air pollution and work towards mitigation of relevant emission sources. Such cooperation will yield significant benefits for all regions, upwind or downwind, and will help decrease the adverse effects of air pollution at the local, regional, and global scales.

Appendix A

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