

## **Hemispheric Transport of Air Pollution 2010 Executive Summary**

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### **Introduction**

The Task Force on Hemispheric Transport of Air Pollution (TF HTAP) was created by the Convention on Long-range Transboundary Air Pollution (LRTAP Convention) in December 2004 to improve the understanding of the intercontinental transport of air pollutants across the Northern Hemisphere. Under the leadership of the European Union and the United States, the TF HTAP has organized a series of projects and collaborative experiments designed to advance our scientific understanding of the intercontinental transport of air pollution, as well as 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 intensively cooperated with the LRTAP Convention centres and task forces and other relevant international and regional organisations. This report, produced by the task force chairs, briefly summarizes the findings and recommendations of a multi-volume assessment produced by the TF HTAP reviewing the state-of-the-science with respect to the intercontinental transport of ozone (O<sub>3</sub>), particulate matter (PM), mercury (Hg), and persistent organic pollutants (POPs)<sup>1</sup>.

### **General Findings**

O<sub>3</sub>, PM, Hg, and 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 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. In most cases, mitigating local or regional emission sources is the most efficient approach to mitigating local and regional impacts of these pollutants. However, without further international cooperation to mitigate intercontinental flows of air pollution, many nations are not able to meet their own goals and objectives for protecting public health and environmental quality. With changing global future emissions, it is likely that over the next 20 to 40 years it will become even more difficult for nations to meet their environmental policy objectives without further cooperation. Cooperation to decrease emissions that contribute to intercontinental transport of air pollution has significant benefits for both source and receptor countries.

### **Main Findings on O<sub>3</sub> and PM**

Based on modeling performed for the HTAP intercomparison, current annual average surface O<sub>3</sub> levels in the northern mid-latitudes are about 35 ppbv. Of this, about 20-25%

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<sup>1</sup> To be printed in the UNECE Air Pollution Series. Advanced drafts are available on the [www.htap.org](http://www.htap.org)

originates from the stratosphere, and a similar proportion is formed from natural precursor sources. The anthropogenic contribution typically exceeds 50%, and over the continental source regions defined in the HTAP intercomparison about half of this fraction originates from sources over the region itself (8-10 ppbv) and about half is transported from sources outside the region. The relative contribution of each of these four fractions varies widely by location and season. In heavily polluted environments, the fraction attributable to local and regional emissions sources may be much larger.

Models indicate that in the northern mid-latitudes, the contribution from intercontinental transport of O<sub>3</sub> typically peaks in spring and fall, and is smallest during the months when O<sub>3</sub> levels are highest due to the peak in production from local and regional emissions. Over the South Asian region, which is largely dominated by the summer and winter monsoon regime, both the local and regional and the intercontinental influences are largest during the winter monsoon period. Intercontinental contributions to surface O<sub>3</sub> in the Arctic are largest in April through June, with a secondary maximum in October and November. Taken together these results indicate that decreasing local or regional emissions is more effective at decreasing the highest O<sub>3</sub> levels, but that O<sub>3</sub> associated with stratospheric origins, natural emissions sources, and intercontinental transport comprise a significant fraction of tropospheric O<sub>3</sub> that is not within the control of local and regional political jurisdictions.

An increasing trend in regional background O<sub>3</sub> concentrations has been measured consistently at a number of remote sites across the Northern Hemisphere, suggesting an increase by a factor of two or more during the latter half of the 20th century. It is likely that much of this change is due to increases in anthropogenic emissions of O<sub>3</sub> precursors. Within the limits of the measurement records, the increase has been comparable throughout all longitudes, and has occurred in all seasons. More recently, a more rapid increase has taken place downwind of eastern Asia, whereas the increases within the boundary layer of central Europe and North America have slowed down. Measurements clearly show that onshore marine airflow above the marine boundary layer can carry O<sub>3</sub> concentrations that approach or exceed air quality standards and objectives.

PM is comprised of different chemical components, some of which are directly emitted as particles by natural or anthropogenic sources (primary PM) and others have their origin in gases that are transformed through chemical and physical reactions and adsorbed on particles (secondary PM). Both primary and secondary PM play a role in intercontinental transport.

The components of PM can be conceptually separated by origin; volcanic eruptions, vegetation, oceans, wind-blown dust, biomass (vegetation) burning, anthropogenic emissions that have been transported on intercontinental scales as well as from local and regional anthropogenic emission sources. The relative magnitudes of these fractions and their sensitivity to emission changes differ by location, season, year, and chemical component. Primary PM such as mineral dust and black carbon, which are directly emitted into the atmosphere, respond linearly to changes in their emission sources. Secondary PM, such as sulphate and organic aerosols, may have a slight non-linear response to emission changes due to their dependence on chemical processes in the atmosphere.

There is a wide range of surface PM concentrations predicted by current models, reflecting large uncertainties in emissions and atmospheric processes as represented in the models. Despite substantial differences in surface concentrations among the models participating

in the HTAP intercomparison, there is much closer agreement in terms of source attributions on an annual average, region-wide basis.

The contribution of natural emission sources and intercontinental transport of anthropogenic emissions contributes 21-25% of sulphate and 10-18% of the black carbon over East Asia, Europe, and North America. For South Asia, the contribution of natural sources and intercontinental transport of anthropogenic sources is 42% of sulphate and 27% of black carbon, which is primarily related to its position in the vicinity of the European outflow. For particulate organic matter (POM), biomass burning and biogenic sources tend to be the largest sources. Models suggest the contribution of biogenic sources and intercontinental transport from anthropogenic sources to regional POM concentrations ranges from 35% in Europe to 71% in North America. For mineral dust concentrations at the surface, 90% in East Asia, 75% in South Asia, and 85% in Europe are from the deserts over Asia, Middle East, and Africa, respectively. In North America, intercontinental transport contributes roughly 67% of mineral dust concentrations with African dust contributing about 3 times more than Asian dust on an annual average, region-wide basis.

In the Arctic, which has few emission sources, European pollution is the largest contributor to surface concentrations of sulphate, biomass burning in the boreal forests of Eurasia is the largest contributor to surface concentrations of black carbon and POM, and Asia is the largest contributor of surface concentrations of mineral dust.

The contribution of intercontinental transport to total column loading (the integrated amount from the surface up to the top of the atmosphere) tends to be 15 to 25% higher than the contribution to surface concentrations or deposition, indicating the importance of transport above the boundary layer and implying more significant contributions to visibility and radiative forcing impacts.

As is the case with O<sub>3</sub>, annual average, region-wide statistics mask significant variability between seasons and within a given region. The seasonal cycles of aerosol concentration and intercontinental transport vary by chemical component and by region. However, there are substantial differences between the seasonal cycles in surface concentration predicted by models participating in the HTAP intercomparison. The model differences are larger for mineral dust than they are for sulphate, black carbon, and POM, and are larger for the Arctic than for the mid-latitude regions.

The importance of biomass burning and windblown mineral dust as sources of intercontinental transport is supported by the observational evidence from satellites that capture the resulting plumes of PM which are visible traveling across oceans and continents. Satellites can now provide quantitative information on intercontinental PM transport and ground-based lidar networks and mountain top measurement sites provide large continuous data sets that characterize such transport events. Evidence of intercontinental transport is also provided in the form of long term trends in observations of surface concentrations and wet deposition measured on remote islands, which in some cases compare well with the trends in emissions in upwind areas.

The HTAP model intercomparison has provided the first set of comparable estimates of intercontinental source-receptor relationships from multiple models by examining the impacts of 20% emission reductions of relevant anthropogenic pollutants in four model regions,

approximating North America, Europe, South Asia, and East Asia. To quantify the relative importance of emissions changes outside each of these regions as compared to emissions changes inside each of these regions, we defined the “import sensitivity” metric as the sum of the changes in concentration or deposition due to a percentage change in emissions in the three other source regions divided by the concentration or deposition change due to the same percentage change in emissions in its own region. Thus, the import sensitivity is a measure of the relative effectiveness of internationally coordinated emission controls on an intercontinental scale as compared to emission controls in a single region. The import sensitivities for different pollutants considered calculated in the HTAP model intercomparison are tabulated in Annex 1.

For O<sub>3</sub>, the impact of 20% changes in anthropogenic emissions of NO<sub>x</sub>, VOC, CO, SO<sub>2</sub>, and primary PM in one region on surface O<sub>3</sub> in the other regions varies from 0.07 to 0.37 ppbv O<sub>3</sub> on an annual average, region-wide basis, as estimated by the mean of the model ensemble. These values are significant in comparison to the response of surface O<sub>3</sub> to 20% decreases of emissions within the region itself, which vary from 0.82 to 1.26 ppbv. The import sensitivities (shown in Table 1) indicate that in all four of the source/receptor regions, coordinated emissions changes in the three other source regions is at least ~50% as important as the emission changes within the region itself. The import sensitivities are larger for column O<sub>3</sub> than for surface O<sub>3</sub>, and exceed one in Europe, East Asia and South Asia.

The largest source/receptor relationship is the impact of North American emissions on European surface O<sub>3</sub> levels. This is followed by the impact of European emissions on South Asian and East Asian surface O<sub>3</sub>. The annual average impact of East Asian emissions on North American surface O<sub>3</sub> is similar to the impact of North American emissions on East Asian surface O<sub>3</sub>, but with peaks in different seasons. European emissions have the largest influence on Arctic surface O<sub>3</sub> followed by North American emissions.

The intercontinental contributions to O<sub>3</sub> concentrations are most strongly influenced by changes in CH<sub>4</sub>, followed by NO<sub>x</sub>, VOC, and CO, in decreasing order. The combined effect of emission reductions of NO<sub>x</sub>, VOC, and CO was shown to be approximately equal to the sum of the impact of reducing each of these precursors by itself. However, the O<sub>3</sub> response to the multi-pollutant reductions is less than additive in simulations in which aerosols and their precursors were also decreased simultaneously. The O<sub>3</sub> decrease over foreign regions produced by regional reductions in anthropogenic CH<sub>4</sub> emissions was shown to be roughly equivalent to the O<sub>3</sub> decrease from the same percentage reduction of NO<sub>x</sub>, VOC, and CO together. The O<sub>3</sub> response to changes in CH<sub>4</sub> emissions, however, requires about a decade to be fully realized, given the relatively long atmospheric lifetime of CH<sub>4</sub>. As an important greenhouse gas, decreasing CH<sub>4</sub> also has significant direct benefits in terms of climate change mitigation.

The annual average, region-wide O<sub>3</sub> values discussed above mask large seasonal and geographic variability, and large variability across models. Current global O<sub>3</sub> 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<sub>3</sub> and its precursors. However, on finer spatial and temporal scales, significant discrepancies exist between estimates from different models and between model estimates and observations, indicating weaknesses in our representation of processes at the resolution of current models. Furthermore, current global

models are not able to reproduce entirely historic O<sub>3</sub> measurements and observed trends, raising concern about our ability to predict changes in the future.

As part of the HTAP intercomparison, the ensemble of O<sub>3</sub> simulations was compared to O<sub>3</sub> concentrations observed by selected surface networks and sondes (balloons) in North America, Europe, and Asia. At most sites, the model ensemble mean generally captures the observed seasonal O<sub>3</sub> cycle and is close to the observed regional mean surface O<sub>3</sub> 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: underestimating O<sub>3</sub> levels at high altitude sites and overestimating summertime surface O<sub>3</sub> concentrations over Japan and the eastern United States.

The ability of current models to reproduce O<sub>3</sub> 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<sub>3</sub> concentrations in the lower and middle troposphere. For the upper troposphere, the bias is often higher. For some polar sites, there are virtually no correlations between models and O<sub>3</sub> 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<sub>3</sub> concentrations aloft.

In the HTAP intercomparison, the difference between models in the response of O<sub>3</sub> concentrations due to 20% changes in precursor emissions are appreciable (20 to 50% of the ensemble mean response) in both the source region and at distant receptors. Across the northern mid-latitudes, the differences between models are largest for changes in European emissions, with a standard deviation of ~0.2 ppb, which is the same order of magnitude as the mid-latitude annual mean response. The standard deviation of the model responses is also ~0.2 ppb 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<sub>3</sub>. The differences between 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.

For PM, the results of the HTAP intercomparison suggest that a 20% change in anthropogenic emissions in any three world regions combined would achieve between 4% and 18% of the change in annual, region-wide average surface PM concentrations that would result from a 20% decrease in emissions within the fourth region. As tabulated in Annex 1, the import sensitivities are similar for surface deposition of sulphate, reactive nitrogen, and carbonaceous aerosols to those for surface concentrations. The import sensitivities for aerosol column loadings are much larger than for surface concentrations.

Surface PM concentrations generally respond linearly to changing emissions from both local and upwind source regions. However, both sulphate and POM, which are not directly emitted but are formed in the atmosphere through oxidation, can be affected by non-linear systems of chemical reactions. Thus, changes in the intercontinental transport of O<sub>3</sub> and its precursors can affect downwind PM concentrations, underscoring the multifaceted nature of the

consequences of a rising hemispheric background of pollution levels in the northern hemisphere on local pollution levels.

Surface aerosol concentrations in the Arctic respond nearly linearly to emission changes in other regions and are most sensitive to emission changes in Europe. However, total column loadings of aerosols 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.

Many of the same limitations and sources of uncertainty faced in estimating O<sub>3</sub> concentrations apply to current models for PM, although there are additional challenges as well. In the HTAP intercomparison, the range of predicted concentrations across the participating models varies across the chemical components of PM. The differences between models is 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 mineral dust surface concentrations. For the Arctic region, where PM levels are very low, the relative spread of modeled concentration estimates is much greater. A detailed comparison between 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 PM observations has not been conducted, however available observational data to support such an evaluation are being compiled and some comparisons have been conducted for sulphate wet deposition.

There is ample experimental evidence from human and animal studies that exposure to ambient O<sub>3</sub> and PM concentrations causes adverse health effects which range from minor sensory irritation to premature death. O<sub>3</sub> and PM also cause damage to a variety of different materials and ecosystems including crops, forests and grasslands, which have important implications for productivity, biodiversity, and food security. O<sub>3</sub> and one of its main precursors, CH<sub>4</sub>, are greenhouse gases that significantly contribute to global climate change.

The highest concentrations of O<sub>3</sub> are typically associated with stagnant conditions, when the contribution from transport is low and the contribution of local and regional sources are most important. However, intercontinental transport has increased baseline O<sub>3</sub> concentrations to the point where they exceed thresholds for protection of vegetation regularly 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 exceedances of such standards will continue to increase.

Relatively few studies have tried to quantify the human health impacts of intercontinental transport of O<sub>3</sub> specifically. Those studies have focused on the impact on the relationship between annual average concentrations and premature mortality. These studies suggest that intercontinental transport can contribute significantly to health impacts of air pollution within a given receptor region. For O<sub>3</sub>, one study based on the HTAP multi-model experiments estimated that intercontinental transport of O<sub>3</sub> contributes 20% to >50% of O<sub>3</sub>-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. Three studies of O<sub>3</sub> transport have suggested that emission reductions in North America and Europe will avoid more mortalities outside these source regions than within the regions themselves. Several modelling studies have estimated that crop yield losses associated with the intercontinental transport of O<sub>3</sub> may be as high as 40% depending on the location, crop, and response function used. O<sub>3</sub> causes tens of billions of dollars in crop damage globally and may be an important contributing factor to the yield gap that currently exists across much of Asia. However, there is significant uncertainty in these estimates.

O<sub>3</sub> contributes significantly to climate forcing, as a greenhouse gas that causes warming directly and indirectly by damaging plants inhibiting their natural uptake of CO<sub>2</sub>. Among O<sub>3</sub> precursors, widespread reductions in emissions of CH<sub>4</sub>, CO, and VOCs better reduce net climate forcing than reducing NO<sub>x</sub>, which may increase forcing over decadal time scales. Decreasing emissions of CH<sub>4</sub>, which is a greenhouse gas itself, will result in near-term decreases in the direct forcing from CH<sub>4</sub> and the direct and indirect forcing of O<sub>3</sub>.

For PM, the experimental and epidemiological evidence for effects on mortality is stronger than it is for O<sub>3</sub>. PM also damages a variety of different ecosystems including crops, forests and grasslands through the processes acidification, eutrophication, and alteration of solar radiation. As in the case of O<sub>3</sub>, these ecosystem impacts have important implications for ecosystem productivity, biodiversity, and food security. PM also significantly influences climate change on regional and global scales, contributing to net warming or cooling depending on its chemical composition and location.

Intercontinental transport events associated with forest fires or dust storms do produce exceedances of short-term PM public health standards (e.g., the impact of Saharan dust events on fine particle 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.

The few studies made so far 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. According to one study, intercontinental transport of PM may be responsible for 380,000 premature mortalities per year on a global basis with a relative 95% confidence interval of 18% to 240% of the central estimate. Most of the impact is attributed to transported mineral dust; those attributed to anthropogenic PM are estimated to be ~25% of the total. As is the case with O<sub>3</sub>, the sum of the health impacts of transported PM pollution in downwind foreign regions can be larger than the health impacts of emissions in the source region itself.

PM is a significant contributor to climate forcing and 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 (BC), CH<sub>4</sub>, CO, and NMVOCs are estimated to have caused a climate forcing since 1750 roughly as large as that from anthropogenic CO<sub>2</sub>. Reductions in PM would improve air quality, but for cooling aerosols, including sulphate, nitrate and POM, this would generally increase warming. Reductions in BC would typically benefit both air quality and climate.

As with O<sub>3</sub>, the climate forcing resulting from changes in emissions of PM and its precursors depend strongly on location, timing, and the background composition. Based on the HTAP intercomparison, the import sensitivity 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 import sensitivities ranging from 10% to 40%. 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 sum of sulphate, black carbon, and POM, South Asia is most strongly influenced by foreign sources, and Europe is the least. The variability between models is very large, highlighting the significant uncertainties in modeling aerosol processes and transport.

The Arctic is experiencing rapid climate change. Arctic climate is affected by O<sub>3</sub> and PM that are transported into the Arctic from other regions, as well as by the climate forcing of O<sub>3</sub> and PM outside of the Arctic. Deposition of black carbon in snow is understood to be an important positive forcing (warming) in the Arctic.

The significance of intercontinental transport may change in the future due to changes in the magnitude and spatial distribution of anthropogenic emissions. The implications for intercontinental transport of changes in anthropogenic emissions and climate have been considered for O<sub>3</sub> and PM by considering a set of global emission scenarios developed for the IPCC (Reference 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<sub>3</sub> and PM precursors will decline, up to 76% for SO<sub>2</sub> 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<sub>x</sub> 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<sub>x</sub> 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. The impact of this redistribution of future emissions and expected changes in future global CH<sub>4</sub> concentrations as specified by the RCP scenarios was explored using linear approximations of the intercontinental source-receptor sensitivities determined under current conditions. The import sensitivities, tabulated in Annex 1, were shown to vary with the expected changes in regional emissions. For North America O<sub>3</sub> concentrations, the importance of emissions changes outside the region relative to emissions changes within the region is expected to double before 2050 under all four RCP scenarios. This suggests that in the future, changes in emissions of O<sub>3</sub> precursors outside the region may be as important as changes within the region. For Europe, the import sensitivity for O<sub>3</sub> increases relatively little under the highest and lowest scenarios. For East Asia, the import sensitivity decreases under a high emission scenario, under which emissions within the region increase, and increases under a low emission scenario, under which emissions in the region decline. For South Asia, the import sensitivity for O<sub>3</sub> declines under both the low and high scenarios. Note that the sensitivity to changes in geographical distribution of emissions within a particular region has not been adequately assessed yet.

Using the linear approximation, the influence of changing CH<sub>4</sub> 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. Under future scenarios, expected changes in CH<sub>4</sub> concentrations have a large influence on surface O<sub>3</sub> changes, in some cases offsetting significant changes in O<sub>3</sub> formation associated with local and regional emissions.

The HTAP model intercomparison also examined the potential impacts of changes in meteorology and transport patterns expected as a result of climate change. Future changes in climate are expected to increase the effect of 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 by changes in atmospheric chemistry and not by changes in transport patterns. The effect of natural emission changes and wider climate-related feedbacks have not been evaluated fully yet.

### **Main Findings for Hg**

Hg differs from other major atmospheric pollutants (e.g. O<sub>3</sub> 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 impacts are 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 other freshwater and marine fish-eating wildlife.

Hg is emitted into the atmosphere as elemental Hg<sup>0</sup> in gaseous form or as oxidized, ionic Hg<sup>2+</sup> in gaseous or particle forms. Hg<sup>0</sup>, 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 deposition and wet removal processes. For purposes of apportioning deposition and subsequent impacts back to emission sources, Hg deposition can be divided into Hg that was released originally from natural sources; legacy Hg that was released from anthropogenic sources originally, was deposited, and subsequently re-emitted; newly-released Hg that was released from anthropogenic sources primarily as Hg<sup>0</sup> and transported on intercontinental scales; and newly-released Hg that was released from local and regional anthropogenic sources primarily as ionic Hg and quickly deposited.

Based on models participating in the HTAP intercomparison, natural and re-emitted Hg account for between 35 and 70% of total Hg deposition on region-wide, annual average depending on the region, and intercontinental transport of newly-released anthropogenic Hg emissions accounts for between 10-30% of total Hg deposition, on an annual globally-averaged basis. East Asia, which accounted for almost 40% of total global newly-released mercury in 2000, is the most dominant of the four HTAP source regions contributing 10-14% of annual Hg deposition 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.

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 when Hg<sup>0</sup> that has built up over the dark winter is rapidly oxidized and deposited in atmospheric mercury depletion events (AMDEs) during polar sunrise.

The long atmospheric lifetime of Hg<sup>0</sup> results in baseline concentrations of 1.5 to 1.7 ng m<sup>-3</sup> Hg<sup>0</sup> in the Northern Hemisphere and 1.1 to 1.3 ng m<sup>-3</sup> Hg<sup>0</sup> in the Southern Hemisphere (at sea level). This inter-hemispheric gradient has been observed consistently in ship-borne observations and is reproduced by current global models. Intercontinental transport of Hg has been observed in episodic events of elevated Hg<sup>0</sup> concentrations recorded at remote mountain top sites and during aircraft measurement campaigns. 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 firm air samples, and suggest that Hg deposition has increased about threefold since pre-industrial times. Decreasing deposition trends observed in Europe and North America are consistent with regional emission controls; however, global trends in concentrations and deposition are ambiguous and may indicate off-setting effects between emission trends in Asia and the other parts of the world, and significant recycling of Hg between environmental components.

The global Hg models participating in the HTAP intercomparison provided relatively consistent estimates of the impact of one source region on another despite significant differences in emissions and chemistry in each model. The import sensitivities, based on 20% emission reductions in the regions and tabulated in Annex 1, suggest that emission controls within East Asia are much more important for deposition levels in that region than are changes in intercontinental transport into the region. For Europe and South Asia, emission changes within the region are most effective at changing deposition levels, but changes in intercontinental transport can result in relatively large changes in deposition within those regions. For North America, emission reductions in East Asia, Europe, and South Asia decrease region-wide deposition more than emissions reductions within North America. In the Arctic, Hg deposition could be the most efficiently controlled by emission reduction in East Asia and Europe due to their proximity to the Arctic.

Current models of global Hg transport reproduce the observed surface Hg<sup>0</sup> concentrations to within 20% of the sparse observations that are available. The models estimate Hg wet deposition to within 80% of the available observations for Northern Hemisphere mid-latitude regions and to within 120% for the Arctic. The level of agreement of current models with available observations, however, masks significant differences between models in the assumptions used concerning emission rates for natural or re-emitted mercury, the oxidation pathways for Hg<sup>0</sup>, and dry deposition fluxes, which are not well understood.

To understand, the main impact that international emissions controls will have on MeHg exposure over intercontinental scales, it is necessary to understand the overall chain of human exposure, including the linkages between atmospheric and oceanic Hg transport, Hg methylation in marine and freshwater ecosystems, exposure to and biomagnification of MeHg in freshwater and marine fish, the international trade of fish, and fish consumption patterns. These linkages are poorly quantified at the present.

Several recent studies have developed global emission projections for anthropogenic Hg emissions in the years 2020 and 2050. Both studies conclude that significant increases in global

Hg emissions, up to 25% in 2020 and 100% in 2050 as compared to 2005, can be expected if no major changes in emission controls are introduced. However, both studies also conclude that the implementation of known emissions control technology could maintain or decrease global Hg emissions. 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 recirculating in the environment by decreasing Hg emissions globally.

The impacts of climate change on intercontinental transport of Hg were not explicitly addressed in the HTAP intercomparison, 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.

### **Main Findings for POPs**

By definition, POPs have long lifetimes in the environment, often cycling between 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 between 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 programs provide good spatial coverage of atmospheric concentration information for most POPs in the UNECE region. However, only a few monitoring programs also analyze POPs in precipitation from which total deposition can be estimated. Some long-term air monitoring programs have observed decreasing concentration trends for some POPs subject to international emissions controls (e.g., technical HCH). For other POPs (e.g., p,p'-DDT at the Arctic stations of Alert and Pallas), observed trends show slow or no significant decline in air concentrations in response to international emissions controls.

Most POPs do not have natural sources, so observed POPs concentrations or deposition can be apportioned into re-emitted POPs that were originally released from anthropogenic sources, newly-released POPs that were released from anthropogenic sources and have been transported on intercontinental scales, and 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. Apportioning re-emitted POPs back to their original sources requires estimating historical emissions and simulating the behavior of POPs in other environmental media over long time periods. As primary emissions are decreased or ceased as a result of emission control measures, re-emission becomes increasingly important.

As part of the HTAP intercomparison, the transport of several POPs (PCB-28, PCB-153, PCB-180, and  $\alpha$ -HCH) covering a range of different physical and chemical properties was simulated by three models using different approaches to describe pollutant transport through environment compartments and different spatial resolutions. Despite their differences, the models provide estimates of annual average atmospheric concentrations, the major transport pathways of selected POPs, and the response to primary emission changes that are within a factor of two to three of the ensemble mean.

Differences in the estimated import sensitivities, tabulated in Annex 1, are primarily a function of the uneven spatial distribution of emissions between regions and the transport characteristics of the pollutant. The largest effect on intercontinental transport is seen for 20% reductions of new  $\alpha$ -HCH emissions in South Asia which decreases concentrations in other regions from 2% to 6%. Decreases in PCB emissions in Europe produce similar levels of response across the other regions. The lowest response is produced by the reduction of  $\alpha$ -HCH in East Asia and North America, and for PCBs in East Asia, due to their relatively low contributions to the total 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  $\alpha$ -HCH).

The regional differences in the efficiency of atmospheric POPs transport alone was 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.

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 modeled and evaluated include PCB-28, PCB-153, PCB-180, and  $\alpha$ -HCH. 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 modeling approaches.

As with Hg, POPs models must not only simulate the behavior 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 is limited, making it difficult to evaluate models and characterize uncertainties.

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. The traditional diet of indigenous Arctic people, which 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 results 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 PBDEs, PFOS, and 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.

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, shifts 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 and the Stockholm Convention is expected to decrease emissions by more than 90% for HCB and PCB, more than 60% for 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 in a variety of ways. Similar to Hg, climate change may significantly alter the exchange of POPs between 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. 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.

## **Main Recommendations**

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 emissions controls on pollution levels and their environmental or public health impacts is limited.

To better quantify the impacts of intercontinental flows of air pollution and the effects of international controls, additional efforts are needed to improve further 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. A program of monitoring, research, and analysis activities is needed that is

- Intentional, making source attribution on global to intercontinental scales an explicit objective of on-going and future efforts
- Innovative, employing novel techniques and developing new methods where needed
- Integrated, generating new insights by combining and comparing information within disciplines and across disciplines, as well as across different pollutants
- Inclusive, engaging a broader community of scientists and air quality management officials in developed and developing countries throughout the Northern Hemisphere.
- Supported by institutions and information networks, fostering and facilitating cooperation between experts within and across disciplinary and community boundaries

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 between continental source regions also need to be assessed further. In future work, the implications of sector-based control strategies in different regions 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. The Stockholm Convention on POPs and the recently opened negotiations under UNEP on a global instrument on Hg provide forums for furthering global cooperation to mitigate sources of POPs and Hg.

For O<sub>3</sub> and PM, however, there is no such global agreement although several regional agreements, in varying states of evolution, address at least some of the sources of transported O<sub>3</sub> and PM. A range of approaches have been suggested for establishing global or hemispheric scale cooperation on mitigation of O<sub>3</sub> and PM, including a global confederation between existing regional efforts building on existing organizational infrastructures and intergovernmental relationships. Such a confederation could facilitate information exchange, capacity building, and technology transfer between regions and greatly facilitate connections between the regional air quality agreements and other existing global agreements, global institutions, and the global scientific community. Thus, a global confederation of regional cooperative programs on air pollution 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 programs appropriate for their circumstances. Such a confederation may be best organized under an existing global institution, such as UNEP, WMO, or a joint UNEP-WMO venture, as in the case of the IPCC.

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. Irrespective of the future role of the TF HTAP the potential for a more

formal global confederation of regional cooperative programs on transboundary air pollution should be explored.

### Annex 1: Import Sensitivities Calculated from HTAP Intercomparison

Import sensitivity is defined as the sum of the changes in concentration or deposition due to a percentage change in emissions in the three other source regions divided by the concentration or deposition change due to the same percentage change in emissions in its own region. Thus, the import sensitivity is a measure of the relative effectiveness of internationally coordinated emission controls on an intercontinental scale as compared to emission controls in a single region.

Scenario	Pollutant	Parameter	North America	Receptor Region		
				Europe	South Asia	East Asia
2001 Ozone		Surface Concentration	46%	74%	46%	66%
2001 Ozone		Total Column	64%	161%	62%	148%
2001 Particulate Matter		Surface Concentration	7%	5%	25%	10%
2001 Sulfate		Deposition	9%	10%	32%	13%
2001 Total Sulfur		Deposition	3%	3%	19%	5%
2001 Black Carbon		Deposition	4%	1%	13%	3%
2001 Particulate Organic Matter		Deposition	3%	1%	12%	4%
2001 Reactive Nitrogen		Deposition	3%	4%	15%	8%
2001 Sulfate		Total Column	33%	33%	59%	31%
2001 Black Carbon		Total Column	32%	21%	19%	17%
2001 Particulate Organic Matter		Total Column	30%	27%	13%	24%
2001 Mercury		Deposition	155%	54%	74%	11%
2001 a-HCH		Deposition	447%	9%	6%	170%
2001 PCB-28		Deposition	10%	3%	91%	129%
2001 PCB-153		Deposition	13%	3%	46%	99%
2001 PCB-180		Deposition	12%	4%	56%	128%
2001a Ozone		Surface Concentration	47%	72%	49%	66%
2030b Ozone		Surface Concentration	112%	82%	25%	42%
2050c Ozone		Surface Concentration	98%	77%	16%	69%

a Calculated from linear approximations of a subset of models participating in the HTAP intercomparison used in scenario comparison

b RCP 8.5, high emissions scenario

c RCP 2.6, low emissions scenario

