

## TF HTAP 2007 Interim Report

**Author Lists:** Coordinating Lead Authors are underlined, Lead Authors are listed in regular text, invited Lead Authors who have not confirmed their participation are listed in *italics*.)

### Chapter 0. Executive Summary (Terry Keating and André Zuber), 1 page

This summary will be drafted as the Task Force's contribution to the main report of the Gothenburg protocol (GP) review. The summary will be drafted after the other chapters are completed, drawing heavily upon the conclusions and recommendations chapter. The summary will address the key tasks of "understanding the hemispheric transport of air pollutants" and "quantify the transcontinental influence". The conclusions should not be "binding" or preclude the further work to be performed for the 2009 assessment. The TF should attempt to summarize the consensus view as to whether intercontinental transport of ozone, fine particles, etc and their precursors (including NO<sub>x</sub>, CH<sub>4</sub>, CO, SO<sub>x</sub>, and organic compounds) is significant in terms of exceedances of policy objectives and what the key uncertainties are. Strictly speaking, the report only has a bearing on the review of the Gothenburg Protocol, therefore, mercury and POPs will not be addressed explicitly.

### Chapter 1. Introduction (Terry Keating and André Zuber), 1-2 pages

This very brief chapter will introduce the Task Force in the context of LRTAP Convention and the purpose and organization of the interim report.

### Chapter 2. Conceptual Overview of Hemispheric or Intercontinental Transport Processes (O. R. Cooper, A. Stohl, R. Doherty, P. Grennfelt, P. Hess), 6 pages

This chapter will introduce the issue of hemispheric or intercontinental transport and the approaches to assess intercontinental transport using an integrated approach to systematic observations, process studies (including short term intensive observations) and modeling.

#### 2.1 Major emissions regions

Very briefly (one paragraph) describe the major population centers and anthropogenic emissions regions of the world. Emphasize eastern Asia and Eastern N. America as major pollutant source regions for the intercontinental transport (ICT) of large plumes due to their relationship to favorable transport patterns, described below. Also describe major biomass burning emission regions, and major dust source regions (northern Africa and eastern Asia). Main concern is with the northern hemisphere due to the far greater emissions. Include a figure showing the major pollutant source regions and the major transport pathways in winter and summer.

#### 2.2 Major types of intercontinental transport processes

**2.2.1 Basic concepts** Winds increase with height; -Isentropes constrain transport in absence of heating; importance of heating at the earth's surface and precipitation; northward motion is generally ascending, southward motion is generally descending, meridional transport much weaker than zonal transport; emphasize difference between tropical and mid-latitude transport process; westerlies at mid-latitudes; easterlies at lower latitudes; there is a transport barrier in the subtropics, so that transport from the midlatitudes to the tropics and subtropics occurs in the boundary layer. ICT of pollutants is dependent of both the timescale of the transport processes and the oxidation/deposition/solubility of the trace gases and particulate matter

**2.2.2 The mid-latitude cyclone airstreams** The Warm Conveyor Belt (WCB) provides rapid high altitude transport, most effective in autumn, winter and spring. Most effective for trace gases that are insoluble with lifetimes greater than 2-3 days. Cold conveyor belt not important for ICT. The dry airstream is an important mechanism for injecting stratospheric ozone into the troposphere. Post cold front air stream can export pollutants to the oceans at low altitude.

**2.2.3 Deep convection** DP followed by upper tropospheric advection, most common in summer. Most effective for trace gases that are insoluble with lifetimes greater than 2-3 days. Can also rapidly loft insoluble gases with short lifetimes like SO<sub>2</sub> before oxidation in the boundary layer.

**2.2.4 Slow, low altitude, zonal-flow** Zonal flow export occurs year round. Transports soluble and insoluble trace gases and particulate matter with lifetimes greater than 1-2 weeks. Imported concentrations are typically quite low.

**2.2.5 Diffuse or small scale boundary layer venting** These events also contribute to ICT but do not produce the large distinct plumes associated with WCBs, widespread organized convective events, or large scale low-level zonal export. Mention orographic lifting, relatively shallow and/or non-precipitating convective lofting of pollutants above the boundary layer.

### **2.3 Impact of intercontinental transport pathways on global and local pollution distributions.**

**2.3.1 Impact of large scale export events** Strong pollution outflow events related to WCBs or widespread organized convective events mainly impact the upper troposphere of downwind continents. To impact the surface, subsidence must occur through anticyclones, isentropic descent behind cold fronts, and convective downdrafts, leading to dilution of the imported pollution.

**2.3.2 WCB and Convective outflow** While the WCB, convective outflow and low altitude zonal transport can produce detectable pollution episodes above downwind continents, much (most) of the pollution exported from a continent will be well mixed with the background pollution and will not produce a distinct pollution signature above a downwind continent.

**2.3.3 Feedbacks between the transported air pollutants and regional climate and meteorology** Atmospheric Brown Clouds; global dimming. Suppression of hurricanes by Saharan dust.

### **2.4 Impact of climate change on future intercontinental transport patterns**

A 1-2 paragraph summary of the relevant studies in the peer-reviewed literature

### **2.5 Outstanding issues and recommendations [for determining the significance of inter-continental transport]**

**2.4.1 Basic transport mechanisms** These are fairly well understood, but work has only just begun on quantifying the export of pollutants by each mechanism.

**2.4.2 Modelling** Additional modeling studies are required to examine the seasonal and interannual variability in ICT.

**2.4.3 Measurements networks** A coordinated measurement network of key trace gases and particulate matter is required along the boundaries of each continent in order to quantify the import and export of the pollutants.

**Chapter 3. Observational Evidence & Capabilities Related to Hemispheric or Intercontinental Transport** (Hajime Akimoto, David Parrish; Surface networks: Kjetil Torseth, Joe Prospero Shiro Hatakeyama, Rich Sheffe; Intensive Observations: Dan Jaffe, Stuart Penkett, Mat Evans, Russ Dickerson; Satellites: David Edwards & Randall Martin, Lorraine Remer, Tony Hollingsworth, Ulrich Platt, John Burrows)

The chapter should focus on carefully selected observational evidence that clearly illustrates the impact of long-range transport on local pollutant levels. The bulk of the chapter should be a clear but concise presentation of the best illustrative examples that we can find to present a balanced, reasonably comprehensive picture of the experimental evidence.

### **3.1 Introduction *Very brief motivation and description of this chapter***

- Very brief motivation and description of capabilities, limitations and needs for the various types of data (surface monitoring, intensive campaigns and satellite)
- Introduction to the “nature” of surface sites:
  - 1) What is meant by regional and global representative sites,
  - 2) Brief historic perspective of how monitoring strategies have evolved,
  - 3) Introduce the difficulties in identifying HTAP events in the boundary layer,
  - 4) Geographic locations which are more suited,
  - 5) Many surface sites lack measurement parameters which makes data interpretation more difficult.
  - 6) Linking boundary layer observations with FT,
  - 7) Need for vertically resolved data in combination with surface sites.
- It would be useful if we could state the usefulness of each data source for specific issues related to the scope of the CLRTAP (e.g. while surface observations of precipitation chemistry are difficult to use to identify the relative contribution originating from intercontinental transport (specific transport events etc), it is probable the longest dataset of consistent data we have for assessing trends, the geographical coverage is good, further these data are the only data representing a flux-term (transboundary fluxes and deposition amounts are key in the Gothenburg protocol revisions), wet scavenging is the major sink term and must be treated properly in the models, they are important as they get the monitoring agency involved in the technical work, etc.) The variability of rainfall events in terms of frequency and quantity coupled with the variability of transport results in a very high statistical variability and consequently great difficulty in resolving trends.

### **3.2 Long-range Transport of Ozone and its Precursors**

*Evidence and illustrative examples from surface measurements, field campaigns and satellites. We should highlight those observations that have most contributed to current knowledge of LRT*

- In-situ surface monitoring of photochemical oxidants and precursors (O<sub>3</sub>, NO<sub>y</sub>, VOCs), in addition vertical profiles from soundings. Data from Convention based monitoring programmes,
- Specific focus on data from “inflow/outflow” sites and sites at high elevations (several recent European projects TROTREP and TOR2 have assessed European ozone trends).
- Asian surface sites including Cape Hedo in Okinawa
- Long-term aircraft programmes (e.g. IAGOS/MOSAIC)
- Asian aircraft missions (PEM-West, Ace-Asia, TRACE-P, APEX, and PEACE; campaigns over China)
- Evidence for hemispheric/intercontinental transport of ozone and precursors from intensive campaigns:
  - across the Pacific (PHOBEA, TRACE-P, ITCT, INTEX, others?).
  - across the Atlantic (NARE, ICARTT, ITOP, others?).
  - across the Eurasian continents (APARE, TRACE-P, ACE-Asia, PEACAMPOT, others?).

- Evidence for hemispheric/intercontinental transport of ozone and precursors from satellite data sets
  - O<sub>3</sub>? (TOMS, GOME, SCIAMACHY?, OMI?....)
  - CO (MOPITT, AIRS...)
  - NO<sub>2</sub>? (GOME, SCIAMACHY?, OMI?....)
- The role of satellite data in intensive field campaigns
  - TRACE-P and INTEX experience, others?

#### *Capabilities, limitations and future needs*

- NO<sub>y</sub> and VOCs data are sparse.
- organic tracers? Several recent studies have demonstrated LRT of Levoglucosan (eg. Stohl et al 2006)
- Representativeness or adequacy of existing satellite data for characterizing intercontinental transport across the Northern Hemisphere
- Issues of vertical sensitivity and measurement frequency in satellite data sets.
- Challenges in validation of satellite measurements
- Future satellite needs: What are the missing observations and what would be the ideal observing requirements of a future satellite mission(s) to track LRT?
  - Orbits, spatial and temporal measurement resolutions etc.
  - Should we develop a set of standard observational platforms and measurements to enhance data consistency globally and examine trends?
  - Tie-in with work of EMEP CCC, WMO GAW, and the IGACO strategy
  - Outreach to regions beyond the LRTAP Convention.

### **3.3 Long-range Transport of Aerosols and its Precursors**

#### *Evidence and illustrative examples from surface measurements, field campaigns and satellites.*

- In-situ monitoring of major inorganic ions in air (SO<sub>x</sub>, HNO<sub>3</sub>/NO<sub>3</sub>, NH<sub>x</sub>, precipitation chemistry, sea-salts, base cations/mineral dust, data sources
- Convention based monitoring programmes. (IMPROVE, CREATE, EARLINET, AERONET, others).
- SKYNET data from sites located mainly in the Eastern Asia from Mongolia to Thailand as well as in Japan
- Lidar network in the East Asia.
- Oceanic aerosol sampling stations (Prospers et al.) operated from early 1980s and extending to the late 1990s.
- Asian surface sites including Cape Hedo in Okinawa
- Asian aircraft missions (PEM-West, Ace-Asia, TRACE-P, APEX, and PEACE; campaigns over China)
- Evidence for hemispheric/intercontinental transport of aerosols and precursors from intensive campaigns:
  - across the Pacific (PHOBEA, TRACE-P, ITCT, INTEX, others?).
  - across the Atlantic (NARE, ICARTT, ITOP, others?).
  - across the Eurasian continents (APARE, TRACE-P, ACE-Asia, PEACAMPOT, others?).
- Evidence for hemispheric/intercontinental transport of aerosols and precursors from satellite data sets
  - Aerosol (MODIS, TOMS AI?, ...)

#### *Capabilities, limitations and future needs*

- In-situ monitoring of aerosol mass/aerosol chemical composition (size segregated), The latter is available mainly on an occasional basis. What can we learn from size resolved information (making these types of measurements is difficult, tedious, and

expensive). From the standpoint of radiative/optical/remote sensing, a size cut at about 1  $\mu\text{m}$  diameter would be needed. Is there any reason to do PM<sub>2.5</sub>/PM<sub>10</sub>?

- aerosol optical properties/aerosol optical depth, Aerosol integrated light scatter would provide important optical/size information on a continuous basis. This would be good for high time resolution data for modeling of events. An AERONET type device is essential for optical depth and as a measure of vertically-integrated aerosol loading.
- aerosol vertical distribution, LIDAR would be ideal but expensive and troublesome. But maybe over time this will improve.
- BC and OC (These (or some other measure of carbon/organics) is essential BC and OC can be relatively easily obtained by filter combustion/reflectance techniques although here too there are problems of interpretation.
- Given the variability of aerosol (and precipitation) concentrations, if one is to characterize trends, one needs to make measurements over extended time periods.
- From the standpoint of modeling events and linking to remote sensing, you need daily measurements. This can be a huge job if you attempt to do everything on this time scale. The strategy should be to pick a critical subset of measurements that you can do on a daily schedule.

### **3.4 Maximum concentrations seen at downwind receptor locations and implications for surface air quality in those regions.**

This section will discuss episodic events that have lead to high concentrations at the surface of ozone, PM or other key pollutants at inter-continental scale. We will want to emphasize that these large episodes are relatively rare, but can bring high concentrations to the surface.

### **3.5 Observational evidence for attribution of source regions.**

This section will discuss methods that have been used to do source attribution, including meteorological methods (trajectories and their derivatives) and chemical methods (e.g. chemical ratios). We will not discuss Global modeling, since this is covered in a separate section, but I think we should discuss “hybrid” methods, for example Flexpart, or other approaches.

- Inverse modeling of emissions from satellite data sets
- Discussion of the important distinction between episodes vs. increases in background concentrations and the implications for surface air quality?

#### *Capabilities, limitations and future needs*

- Does a sufficient satellite database exist to evaluate the predictions of current models?
- Satellite data are limited in vertical sensitivity, ability to observe diurnal variation in emissions
- Challenges in validation of satellite measurements

### **3.6 Can we track long-term trends in hemispheric transport from existing surface observations?**

This section will describe what evidence exists to assess trends and whether the existing data are adequate to do this.

### **3.7 Concluding Remarks** future needs for observations to understand HTAP

#### **4.1 Introduction**

(What are the requirements of emission inventories for the purposes of the TF?)

#### **4.2 Present-day emission inventories**

(What is available and accessible now for present-day emission inventories; specifically include brief discussion of issues and species relevant for TF, i.e., NO<sub>x</sub> and VOC for O<sub>3</sub>; CO and CH<sub>4</sub>, fine PM, Hg and other trace elements, POPs. Reflect on existing global and regional mechanisms for official reporting and verification of inventories, e.g., under UNFCCC, UNECE, CLRTAP.)

**4.2.1 Global inventories and databases** (GEIA, EDGAR, UNFCC, and GAINS, plus special inventories like Bond et al. [2004] for carbonaceous aerosols and Bouwman et al. [1997] for ammonia)

**4.2.2 Regional and national inventories and databases** (EMEP, RAINS-EUROPE, RAINS-ASIA, TRACE-P, REAS, APINA, and CEPMEIP; other key papers on regional emissions; examples of EI in selected countries representing different states of EI development, e.g., USA, China, India, UK, Russia.)

#### **4.3 Uncertainties and verification of present-day emission inventories**

(How are uncertainties estimated? How can we verify emissions by observation and modeling? What are the implications of current levels of uncertainty?)

**4.3.1 Quantification of uncertainties.** (Methods currently used. What are the estimated uncertainties?)

**4.3.2 Intersection of inventories with observations and modelling** (An introduction to emission inventory issues that bear on Chapter 6: inferences of emissions from field campaigns, inverse modeling, satellites...)

**4.3.3 Important/Uncertain sources** (Anthropogenic biomass burning, aircraft and ships, animals, industrial processes, small-scale combustion, road transport...)

#### **4.4 Projection of future emissions**

(What is the state of knowledge about future emissions?)

**4.4.1 Driving forces** (What are the important factors that determine future emission levels? Discussion of socio-economic forecasts, environmental and trade policies and agreements, technology development ...)

**4.4.2 Methods** (What approaches have been adopted to forecast future emissions? What are their strengths and weaknesses?)

**4.4.3 Future emission inventories** (Examples of available global projections, e.g., IPCC/SRES and RAINS/GAINS, plus other special studies like Rao et al. [2005] for global black carbon, Streets et al. [2004] for global carbonaceous aerosols, Klimont et al. [2001] for Asian air pollutants; discussion of their adequacy for TF needs; the importance of changing spatial distribution of emissions around the world in the future.)

(What have we learned from past experience? Is harmonization of forecasts desirable and feasible?)

**4.5 Natural emissions** How well do we know the various sources of natural emissions? What factors might make natural emissions change in the future? Discuss mineral dust, biogenic VOC, methane, volcanoes, wildfires [and relationship to anthropogenic biomass burning]...

**4.6 Harmonization of different inventories** What have we learned from previous experience? Is harmonization of inventories desirable and feasible? Including discussion forums for emission inventories, such as IPCC, GAP Forum, UNECE-TFEIP, ACCENT, and NARSTO; technical issues for modeling, such as overlapping domains, incomplete coverage, spatial and temporal resolution, speciation [size and chemical] and release height; and the roles of historical inventories, specialized industrial and urban inventories, etc..

**4.7 Recommendations** Improvements that can be made to help the TF in the short-term and in the long-term and prioritization of their importance. Identify opportunities for capacity building and outreach.

## **Chapter 5 Regional, Hemispheric & Global modelling** (Greg Carmichael, Frank Dentener) A para to introduce the chapter objectives

### **5.1 Methods for diagnosing or quantifying transport** (Dick Derwent, Michael Prather, Martin Schultz, Qinbin Li)

**5.1.1 Introduction and definitions** Source contribution, Source attribution, Source apportionment, Source-receptor relationship. Issues of baseline increases vs. AQ peak exceedances

#### **5.1.2. Techniques appropriate to inert tracers or to simple linear systems**

Analyses of observations of dust, haze, PM, CO, PAN, O<sub>3</sub>, POPs, CO<sub>2</sub>, nuclear weapon debris, cosmogenic radionuclides, biomass burning plumes. Back-track trajectory statistics. Lagrangian particle dispersion models. Source foot-prints (Greens' functions)Field-of-view, Tagging experiments for CO, Tagging experiments for stratospheric ozone, Holzer-Hall transport statistics

**5.1.3. Source-receptor relationships in more complex situations** Source-receptor relationships as model constructs – validation against observations, Source-receptor relationships in acid rain systems. Continental chemical inflow and outflow studies. Emission reduction studies, origins of ozone background.. Ozone labelling. Pulse experiments

### **5.2 Estimates of Ozone Transport and its precursors** (Arlene Fiore, Oliver Wild, Carey Jang, David Stevenson, Hiroshi Tanimoto, Don Wuebbles)

**5.2.1 Introduction** Summarize previous studies Table or ranges stated in text (?) with estimates for regions relevant to HTAP, taken from Holloway et al., ES&T, 2003; updated in Fiore et al., EM, 2003; needs to be updated with new literature (2003-2006) and of course any older studies we missed. Description of key uncertainties that will be eliminated through coherent HTAP framework: different (a) study aims (b) methods used (c)regional definitions (d) metrics

**5.2.2 Summary of HTAP multi-model study** Description of objectives Identify sensitivity of ozone concentration and deposition response in receptor regions to 20% decreases in o<sub>3</sub> precursors in source regions. Estimate present-day contribution of source region to concentration/deposition by scaling results to 100% reductions, acknowledging uncertainty associated with non-linearities (e.g. quantitative uncertainty estimates from prior work (Wild et al.; Doherty; others?); expected to be small for CO, NMVOC, and CH<sub>4</sub>, and even NO<sub>x</sub> over

intercontinental distances) Use differences across models to provide a measure of uncertainty. Adopt a coherent cross-model framework to isolate uncertainty associated with combination of model differences in emissions, transport, loss processes. Description of methods: 3D global tropospheric chemistry models at 4x5 resolution or finer. refer to Figure of S-R regions (introduced in 5a or earlier chapter?). 2001 met year. individual models use their best emissions estimates for 2001. Define metrics to be examined – decisions needed here (discuss in 5a?). Choose one or two regional mean statistics that could be included in traditional S-R tables and easily normalized to emission changes in source region, as well as one statistic that would give some measure of event variability. Some possibilities: Annual/seasonal/monthly of straight mean / daily max 8 hr / AOT40 / SUM06 for ozone straight mean for O<sub>3</sub> (+ NO<sub>y</sub>?) deposition.

**5.2.3 Present-Day S-R relationships for O<sub>3</sub>** Discuss response of chosen metrics to NO<sub>x</sub>, NMVOC, CO, CH<sub>4</sub> Some options: Regional Means: entire region? land-only (for deposition)? Population weighted (for air quality metrics)? Show monthly variation in chosen metrics (mean + sdevs) for each S-R pair? Episodic events: probability distributions of daily mean/daily max 8hr/hourly o<sub>3</sub>? Temporal standard deviations of o<sub>3</sub>? Frequency of events above threshold values relevant to standards – bar plots? Scale results up to total contribution to give range of present-day estimates. Uncertainties in S-R Discuss full model range here – coherent results across models? Mean results more certain than episodic / exceedance type statistics? Normalize results by applied emission perturbation -- results more coherent? Any obvious dependence on spatial resolution? Are some metrics more uncertain due to spatial resolution? Are any processes clearly identified as most important in contributing to uncertainty (cf TP1)? Is there a simple relationship; strong non-linearities? Can we address this by looking at absolute emission changes and absolute responses, which will not be exactly the same across models due to different inventories? Any way to assess minimum level of certainty needed for useful estimates? Role of meteorological variability (from literature)?

**5.2.4 Future changes in S-R relationships** ACCENT/PHOTOCOMP results. Changes due to changing balance of emissions can be discussed in light of various SR simulations. Changes in future background resulting from changes under specific emission scenarios in previous studies (e.g. ACCENT, Szopa 2006 for 2030 CLE/MFR/etc in Europe; Fiore 2002 A1b in USA; others?) Changes due to climate change – Murazaki and Hess; Stevenson; others?

**5.2.5 Priorities for further study** Highlight gaps in understanding that can be filled with additional model studies (resolution; future impacts; improved process treatment) that would reduce uncertainty.

### **5.3 Aerosols [and their precursors]** (Michael Schulz, Rokjin Park Christiane Textor, Isabelle Bey, Dorothy Koch, Mian Chin)

**5.3.1 Introduction** Motivation: Why may short-lived species, such as aerosols, be transported on hemispheric scales? Observed evidence of long range transported “aerosol clouds” Role of aerosol ageing and secondary aerosol formation for export efficiency. Role of aerosol size&solubility and climate for export efficiency. Role of dust and seasalt as episodic contributors to PM levels Review model studies on intercontinental aerosol transport

(pollution and natural). Introduce to specific goals of HTAP work on transport of aerosols and their precursors: Estimate the combined effect of a continent wide reduction of anthropogenic emissions for PM levels on other continents and their regions. Evaluate the available aerosol models and any combination of these to obtain a best estimate and a range of uncertainty of such an effect Explore the role of aerosol processes responsible for non-linear relationships between emissions and PM levels. Determine impact for confidence in simulating future air pollution scenarios Identify ways to improve the aerosol models .

**5.3.2 Overview on available models** Aerosol modeling concepts, required complexity for simulating PM levels, review of previous model intercomparisons of regional and global models, review spatial resolution and boundary conditions, emphasis on models participating in HTAP experiments, role of regional and global aerosol models for hemispheric transport assessment

**5.3.3 Model simulations of hemispheric transport** Review of published work. Introduction to HTAP experiments Documentation of impact of delta 20% experiments (SR1 plus SR6 combined eventually with TP1) on PM levels in remote continents Discussion of anthropogenic and natural aerosol contributions Differentiation of primary and secondary aerosols Documentation of seasonality of transport

**5.3.4 Evaluation of aerosol transport model results** PM evaluation, per continent. Aerosol composition and size. AOD evaluation, AeroNet based. Discussion of AOD to be used for particulate air pollution assessment Deposition evaluation, per continent – seasonality and contribution of long range transported component Explore validation of wet deposition parameterizations. Vertical profile documentation models, aircraft measurements and Calipso. Recommendations for standard evaluation procedures. Interhemispheric transport efficiency Documentation of export efficiency for different regions. Why are the aerosol models different with respect to transport? Role of photo-oxidant availability for transport efficiency of aerosol precursors. Identification of aerosol processes which can be suspected to introduce non-linear relationships between emissions and PM levels

**5.3.5 Impact of climate change on aerosol transport** Review of publications Role of changed hydrological cycle for aerosol life times Indirect effects impacting secondary aerosol formation

**5.4.6 Summary and Recommendations** Summary with respect to policy relevant questions. Which model experiments are suggested to improve confidence (other diagnostics, sensitivity studies, other scenarios, other models)? What is proposed to improve estimates of aerosol transport (model improvement, assimilation, observational data basis, multi-model ensembles)?

## **6. Integration of observations, modeling, and emissions** ([Daniel J. Jacob](#), [Len Barrie](#), Oystein Hov, Rudy Husar, Brendan Kelly, Jill Engel-Cox)

### **6.1 Introduction**

Quantitative assessment of intercontinental transport of pollution in terms of source-receptor relationships requires chemical transport models (CTMs) driven by best

estimates of emissions. These CTMs need to be constrained and evaluated by atmospheric observations, and in turn provide information on what kind of observations are most needed for model testing. This exchange of information between observations and models defines an integrated observing system.

### **6.2 Observing system concept**

Describe Bayesian optimization of information from emissions, CTMs, and observations as an observing system to improve understanding of intercontinental transport and quantify source-receptor relationships. Point out that separation of model and observational approaches is artificial – one should think rather of an interactive partnership. Show optimization flow chart starting from a priori knowledge of emissions and transport to drive a CTM (forward model), followed by comparison to available observations and use of this comparison to iteratively improve the model and the observation network; and finishing with inference of top-down information to improve the a priori knowledge.

### **6.3 Past applications of observing system concept**

Review continental outflow and intercontinental transport analyses from TRACE-P, ICARTT, INTEX-B missions (anything else?). Critical role of aircraft as integrator of information for transport on intercontinental scales. Use of models as transfer functions between different observation types.

### **6.4 Future applications**

Integration of satellites, aircraft, and models during the POLARCAT mission to the Arctic in 2008. Exploitation of data from satellites already in space (Terra, Aqua, Aura, GOME, Envisat, GOME-2) for data assimilation and inverse modeling.

### **6.5 Development needs**

Satellites in sentinel orbit: GEO, L-1. Continuous in situ monitoring using UAVs, background sites. Improved data assimilation capabilities, model adjoints.

### **6.6 Conclusions**

Integration between models and observations is critical to advancing understanding of intercontinental transport of pollution. Importance of taking the observing system perspective. Start by posing the specific questions to answer and design observing system around it. Need for satellites in sentinel orbit, UAVs, advanced inverse modeling tools.