Chapter 2: Conceptual Overview of Hemispheric or Intercontinental Transport Processes

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2.1 Major emissions regions

The magnitude and impact of hemispheric and intercontinental scale transport of pollutants is largely controlled by the global distribution of the human population and anthropogenic emissions. The northern hemisphere contains the great majority of humans (88%) and emissions from fossil fuel combustion, for example 91% of all anthropogenic NO_x emissions (year 2000 EDGAR estimate) [*Olivier and Berdowski*, 2001]. The major emissions regions of the northern hemisphere are located in eastern USA/southeastern Canada, western and central Europe, and southern and eastern Asia. The emissions regions along the east coasts of Asia and North America 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, these emissions are not lofted to the same extent as those on the east coasts of Asia and North America. Instead, the European emissions are exported at relatively low altitudes and have a strong impact on the Arctic [*Stohl et al.*, 2002; *Duncan and Bey*, 2004]. All of these regions commonly experience deep convection during spring and summer which can quickly transport pollutants from the boundary layer to the upper troposphere.

2.2 Major types of intercontinental transport processes

2.2.1 Basic concepts

The term source-receptor relationship is used to assess the contribution of an upwind continent or region to its receptor location [*Wild et al*. 2001/4; *Fiore et al*. 2002; *Derwent et al*., 2004]. Source-receptor relationships can be interpreted as: 1) the direct transport of plumes from the source to the receptor site, and 2) the contribution of the source to the overall background concentrations at a receptor site. While the transport of plumes involves a number of distinct episodic transport events occurring on relatively short time and spatial scales, the background concentration is the result of the cumulative effect of episodic transport events on longer time and spatial scales. Source-receptor relationships are not independent of a constituent's lifetime. The source-receptor relationship of short-lived constituents with lifetimes of a few days is only determined by the rapid and direct transport pathways between the source and the receptor, with the background concentration being essentially zero. In contrast, the source-receptor relationship of long-lived species (e.g., methane- lifetime 8.4 years) will mostly be determined by the contribution of the source to the background component of the distribution.

Basic structural and dynamic aspects of the atmosphere most important for hemispheric pollutant transport are as follows. 1) Mid-latitude zonal mean winds are generally westerly throughout the troposphere, causing intercontinental transport to primarily occur from west to east in the mid-latitudes. In the tropics intercontinental transport is generally from east to west, with easterlies throughout the lower and mid-troposphere. 2) Wind speeds generally increase with height, causing pollutants at higher altitudes to be transported more rapidly. Thus, processes which loft pollutants into the mid- to upper troposphere are most conducive to long-range intercontinental transport. 3) Winds are generally stronger in winter than in summer, causing more rapid intercontinental transport during winter months. 4) Meridional winds are generally weaker than zonal winds, thus pollutants tend to be transported zonally.

Hess [2005] suggests that the troposphere can conveniently be separated into a regime where the air has been predominantly convectively processed (on a timescale of 40 days), and a regime where it has not. In isentropic coordinates this regime boundary occurs between 300 and 310^o K regardless of season or hemisphere. The nonconvective regime roughly corresponds to extratropical regions and is largely dominated by parcel dispersion along isentropic surfaces. These surfaces are generally oriented upward and poleward, therefore, poleward moving air tends to ascend, while equatorward moving air tends to sink. Meridional stirring occurs along isentropic surfaces oriented parallel to the Earth's surface on a timescale of approximately 20 days [*Bowman and Carrie*, 2002; *Bowman and Erukhimova*, 2004]. While dry convection, shallow convection, mountain valley and topographic circulations may play some role in venting the boundary layer in this regime, synoptic venting of the boundary layer through warm conveyor belts is most important [*Merrill and Moody*, 1996; *Stohl and Trickl*, 1999; *Cooper et al*., 2001; *Hess*, 2005]. Synoptically, dispersion along isentropic surfaces can be associated with the passage of cyclonic and anticyclonic mid-latitude systems. Cross isentropic dispersion also occurs as parcels are radiatively cooled or heated, or as latent heat is released.

The convectively influenced regime occurs predominantly within the tropics [*Hess*, 2005]. Here rapid ascending motion occurs mainly within a narrow band in association with the ITCZ; slow descent occurs throughout much of the rest of the regime. Within this regime rapid transport between surface source and receptor sites is not expected. The northern hemisphere summertime mid-latitude mid- and upper-troposphere is also predominantly convectively influenced, with convection being a dominant mechanism for venting the continental boundary layer.

A number of studies have shown semi-permeable barriers to transport between the extra-tropics and the tropics [*Pierrehumbert and Yang*, 1993; *Bowman and Carrie*, 2002; *Bowman and Erukhimova*, 2004; *Hess*, 2005]. Thus, the background concentration at mid-latitude sites is most immediately affected by mid-latitude emissions, with a much slower contribution from tropical sources. The mass exchange between the extratropical and tropical regions is estimated to be 1 – 2% per day of the extratropical mass, with lag times between the northern and southern hemisphere tracer concentrations approaching 2 years [*Bowman and Erukhimova*, 2004]. Equatorward transport between the extra-tropics and tropics occurs primarily in shallow flow near the Earth's surface, often behind cold fronts [*Bowman and Carrie*, 2002; *Bowman and Erukhimova*, 2004]. The return poleward flow occurs in a more diffuse circulation in the upper troposphere.

2.2.2 The mid-latitude cyclone airstreams

Mid-latitude cyclones tracking from west to east are important mechanisms for the export of trace gases and particulate matter from the east coasts of Asia and North America throughout the year, even in summer when these weather systems are weaker [*Merrill and Moody*, 1996; *Cooper et al*., 2002; *Stohl et al.*, 2002]. The cyclones are typically composed of four airstreams that influence trace gas mixing ratios and relationships in the troposphere [*Bethan et al.*, 1998; *Stohl and Trickl*, 1999; *Cooper et al*., 2001]. Three of these airstreams, the warm conveyor belt (WCB), cold conveyor belt (CCB) and dry airstream (DA) produce the distinctive comma cloud of a mature mid-latitude cyclone [*Browning and Monk*, 1982; *Browning and Roberts*, 1994; *Bader et al.* 1995; *Carlson*, 1998]. The WCB is located on the eastern side of the cyclone, ahead of the surface cold front. The air originates at low altitudes in the warm sector of the cyclone and travels poleward, ascending along moist adiabats into the mid- and upper troposphere, above the CCB. The DA, which is associated with stratospheric intrusions, originates at high altitudes in the upper troposphere and lowermost stratosphere on the poleward side of the cyclone and descends isentropically into the mid- and lower troposphere on the polar side of the cold front. The post cold front airstream (PCFA) is the cold, dry, and stable air mass in the lower and mid-troposphere that flows behind the cyclone cold front and beneath the DA [*Cooper et al.*, 2001].

The WCB is the most important airstream for rapid intercontinental pollutant transport because of its ability to loft polluted boundary layer air from the cyclone warm sector to the upper troposphere in the vicinity of the jet stream. Following the mid-latitude cyclone storm tracks, the jet stream then rapidly transports the pollutants downwind. Transport times from the moment North American boundary layer air is lifted within the WCB until it reaches the European free troposphere are typically 3-4 days [*Stohl et al*., 2002; *Eckhardt et al*., 2004], and in some cases less than 2 days [*Stohl et al*., 2003b]. Several additional days are required for the N. American emissions to reach the European surface with the greatest influence over the Mediterranean [*Stohl et al.*, 2002]. The greater distance associated with trans-Pacific transport results in slightly longer transport times, and in some instances requires two WCBs to transport emissions from Asia to N. America [*Cooper et al*., 2004a].

Mid-latitude cyclones can also export pollutants from Asia and North America at low altitudes as discussed in Section 2.2.5, either when the warm sector of the storm pushes offshore and the WCB is too weak to loft the pollutants, or when the cold stable air in the PCFA quickly advects fresher emissions offshore.

2.2.3 Deep convection

Deep convection is triggered when the Earth's surface is sufficiently warmer than the overlying air to produce a conditionally or absolutely unstable atmosphere, such as during daytime over land, or the advection of cooler air masses over a warm ocean surface. Convective circulations encompass small-scale fair weather cumuli, active thunderstorms (cumulonimbus clouds) and mesoscale convective systems [*Cotton et al*., 1995]. The corresponding lifetime of these systems increases with their size from minutes to about half a day. Another weather system shaped by organized deep convection, is the tropical cyclone whose lifetime is on the order of a week. Furthermore, in summertime, over land even the WCBs of extratropical cyclones are characterized by embedded deep convection, and the distinction between deep convection and slant-wise WCB ascent becomes somewhat arbitrary [*Kiley and Fuelberg*, 2006]. An extreme form of convection is the so-called pyro-convection occurring over large forest fires, which can inject large quantities of aerosols and trace gases into the upper troposphere and also deep into the stratosphere [*Fromm et al*., 2000; *Jost et al*., 2004; *Fromm et al*., 2005].

Convection is an important mechanism for vertically transporting air pollutants [*Dickerson et al*., 1987; *Lelieveld and Crutzen*, 1994] out of the boundary layer and into the middle and upper troposphere, where the stronger winds can rapidly transport the pollutants across intercontinental distances. Globally, the mass flux out of the boundary layer due to deep convection is comparable to the mass flux caused by the large-scale slantwise ascent in extratropical cyclones [*Cotton et al*., 1995]. In addition, the ascent in deep convective cells takes only minutes, whereas the ascent in cyclones takes from hours up to two days. For trace gases with a rather short lifetime in the lower troposphere, this has the consequence that they can reach the upper troposphere in convective cells but not with WCBs. For instance, *Speidel et al*. [2007] observed large enhancements of sulfur dioxide in the upper troposphere over Europe, after lifting by deep convection over North America followed by intercontinental transport.

2.2.4 Diffuse or small scale boundary layer venting

Export of pollution from the boundary layer to the free troposphere can occur whenever an air parcel is transferred above the boundary-layer height. Since over land the boundary layer has a distinct daily cycle with a maximum during the day and a minimum during the night, a residual layer is formed upon the transition from day to night [*Stull*, 1988]. This residual layer is decoupled from the surface and experiences higher wind speeds than the air in the boundary layer, particularly when a nocturnal low-level jet is present [*Angevine et al*., 1996]. The residual-layer air can (partly) remain in the free troposphere the next day if the boundary layer is less deep than on the previous day (e.g., upon export from the continent over a cooler ocean surface), or if other vertical transport processes lift it to higher altitudes. Topography has a large influence on vertical pollution transport, as it generates variability in the boundary layer height, and the formation and breaking of gravity waves through a variety of different processes. In particular, the thermal circulations encountered in mountainous regions can trigger vertical lofting. For instance, *Henne et al*. [2004] estimated that under fair weather conditions in summer, three times the valley volume can be lofted into the free troposphere per day. Once pollutants are vented from the boundary layer they are subject to various long-range transport processes discussed above, should their lifetimes be of similar or greater magnitude than the transport times.

2.2.5 Slow, low altitude, zonal flow

Air masses can also be transported over long distances without being lifted. Often, this involves the formation of a residual layer, following the collapse of a daytime boundary layer. It can also happen in a strongly stable atmosphere, where hardly any convective boundary layer forms, for instance at high latitudes in winter. Because dry deposition and the potential for cloud formation are limited under such conditions, aerosols and trace gases can be transported over long distances, even though transport speeds are lower than in the upper troposphere. Arctic Haze [*Barrie*, 1986], which can cover large parts of the Arctic in winter and spring, is often the result of such low-level long-range transport [*Klonecki et al*., 2003; *Stohl*, 2006]. The phenomenon has also been observed downwind of North America, where layers with extremely high concentrations of oxidized nitrogen were found far downwind over the North Atlantic Ocean [*Neuman et al*., 2006]. These layers can even reach the Azores [*Owen et al*., 2006] and probably Europe [*Li et al.* 2002; *Guerova et al.* 2006]. Similar transport pathways have been identified across the Pacific [*Liang et al*., 2004; *Holzer et al*., 2005].

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

2.3.1 Impact of large scale export events

Large scale export events from Asia and N. America have strong episodic impact on the chemical composition of the free troposphere above downwind continents, while their impact on surface sites of downwind continents is less frequent and more dilute. Focusing first on transport from Asia to North America, modeling studies indicate that episodic long-range transport of CO from Asia to the northeastern North Pacific region occurs year-round every 10, 15 and 30 days in the upper, middle and lower troposphere, respectively [*Liang et al*., 2004], with 3-5 Asian plumes impacting the U. S. west coast boundary layer between February and May [*Yienger et al*., 2000]. Aircraft studies have detected strong Asian plumes in the lower and mid-troposphere above the eastern North Pacific [*Heald et al*., 2003; *Nowak et al*., 2004] and the US west coast

[*Jaffe et al.*, 1999; *Jaffe et al.*, 2003a; *Jaeglé et al.*, 2003; *Cooper et al.*, 2004ab] with CO on occasion reaching 300 ppbv. Most of these free tropospheric observations, all with CO in excess of 200 ppbv, were associated with WCB export from Asia, however some low altitude events were associated with export and transport in the lower troposphere. When these plumes have intersected the surface of the Washington State coastline, CO has reached as high as 180 ppbv. These events are not always associated with elevated ozone mixing ratios, and the enhancements are difficult to detect at the surface [*Hudman et al.*, 2004]. As Asian pollution plumes continue travelling east, clear enhancements of ozone and aerosol concentrations can also be found over Europe [*Stohl et al.*, 2007]. Asian pollution can also reach Europe via westward transport with the monsoon circulation from India to Africa and the Mediterranean [*Lawrence et al.*, 2003].

In terms of large scale N. American export events influencing Europe, confirmation of their strong impact has only been made in the free troposphere above Europe [*Stohl and Trickl*, 1999; *Stohl et al.*, 2003a], or at high altitude sites in the Alps [*Huntrieser et al.*, 2005], all involving WCBs. Concerning low altitude surface sites, *Derwent et al.* [1998] found 5 probable cases of N. American emissions influencing Mace Head on the west coast of Ireland, but the pollutant concentrations were quite low. The only major N. American export event to show a strong impact on low altitude European surface sites involved smoke plumes from the widespread biomass burning in Canada that caused CO mixing ratios to reach 175 ppbv at Mace Head during August, 1998 [*Forster et al.*, 2001].

High-latitude Europe and Siberia are cold enough to allow direct transport of air pollution from these regions into the Arctic lower troposphere [*Barrie*, 1986, *Klonecki et al.*, 2003; *Stohl*, 2006], contributing to Arctic Haze, a mixture of sulfate and particulate organic matter and, to a lesser extent, ammonium, nitrate, black carbon, and dust aerosols [*Quinn et al.*, 2007]. Typically, this pollution traverses the Arctic and reaches high-latitude North America [*Sharma et al.*, 2006].

Only a few studies have examined the transport of European pollution to Asia [*Newell and Evans*, 2000; *Pochanart et al.* 2003; *Wild et al.* 2004; Duncan and Bey, 2004]. *Newell and Evans* [2000] estimated that some 25% of the air parcels arriving over Central Asia have previously crossed over Europe, and some 4% have originated in the European boundary layer. *Pochanart et al.* [2003] have shown that average ozone and CO concentrations in East Siberia are enhanced in air masses transported from Europe. Over Japan, impacts of North American and European emissions are similar [*Wild et al.* 2004].

2.3.2 Contribution to background pollution

The combination of long transport times and the mixing of air masses from diverse source regions determines the distribution of background trace gas concentrations. The background concentrations of surface emissions tend to be oriented along isentropic surfaces in the mid-latitudes due to the rapid mixing along these surfaces [*Plumb and McEwan*, 1978; *Bowman and Carrie*, 2002; *Bowman and Erukhimova*, 2004; *Hess*, 2005]. The atmospheric circulation tends to transport the highest concentrations of constituents towards the poles [*Hess*, 2005]. Recent studies suggest background levels of pollutants along the west coast of North America [*Jaffe et al*. 2003b; *Parrish et al*., 2004] and Europe [*Derwent* et al.1998; *Oltmans et al*., 2006] are changing. *Fiore et al.* [2002] found an out-of-phase relationship between background and locally produced ozone at the surface of the U.S. in summer. During high pollution episodes under stagnant conditions background ozone concentrations were small; when the boundary layer was more thoroughly vented locally produced ozone was at a minimum, but background ozone concentrations were greater.

2.3.3 Feedbacks between transported air pollutants and regional climate and meteorology

Trace gases and aerosols have been shown to modify both the radiational balance of the atmosphere as well as cloud formation and precipitation. Therefore it might be expected that pollutants from an upwind source can modify the climate and meteorology of a receptor site, which may then create feedback mechanisms that modify how pollutants are transported around the globe. Studies are needed to quantify these effects.

Long-range transport of aerosols has received much attention due to the discovery of atmospheric brown clouds [*Ramanathan et al. 2001*a], which form a haze layer over much of southern Asia between December and April. Regionally, the absorbing haze (mainly black carbon) decreased the surface solar radiation by an amount comparable to 50% of the total ocean heat flux and nearly doubled the lower tropospheric solar heating [*Ramanathan et al.*, 2001b]. The seasonality of brown clouds and their dimming effect is currently under investigation as part of Project Brown Cloud [*Ramana et al.*, 2006].

The efficiency of sunlight absorption in aerosol layers is greater in the Arctic than at lower latitudes, due to the prevalence of large solar zenith angles, and the high albedo of snow and ice. Black carbon is a minor but important component of the Arctic Haze and causes heating of the haze layers [*Quinn et al.*, 2007]. In addition, deposition of black carbon onto snow and ice results in a reduction of the albedo [*Warren and Wiscombe*, 1980; *Clarke and Noone*, 1985]. It has been suggested that the climate forcing due to this albedo effect is significant compared to the effect of greenhouse gases [*Hansen and Nazarenko*, 2004].

Intercontinental transport of desert dust from Asia and North Africa has been well documented [e.g. *Liu et al.*, 2003a]. When the dust-laden Saharan Air Layer advects from Africa to the tropical North Atlantic it can engulf tropical waves, or pre-existing tropical cyclones, and the dry air, temperature inversion, and strong vertical wind shear (associated with the mid-level easterly jet) may inhibit their ability to strengthen and suppress hurricane formation [*Dunion and Velden*, 2004]. *Evan et al.* [2006] demonstrate a strong relationship between North Atlantic tropical cyclone activity and atmospheric dust cover in support of this hypothesis.

The radiation budget of the atmosphere is sensitive to the vertical ozone distribution, which is controlled by transport. Tropospheric ozone has caused enhanced warming (>0.5 C) over polluted northern boreal zones during summer between 1890 and 1990, and between 0.4 and 0.5 C during winter and spring, between 1890 and 1990 [*Shindell et al.,* 2003].

2.4 Impact of climate change on future intercontinental transport patterns

Climate change will affect intercontinental transport (ICT) patterns through alteration of the large-scale circulation and regional climate and hence the transport processes described above. Predictions of changes in regional climate are generally less robust than large scale changes [*Giorgi et al*., 2001]. Examining experiments generated for the IPCC 4th assessment report, *Held and Soden* [2006] found a number of changes robust across the models: a decrease in deep convective mass flux**,** and an enhancement in patterns of evaporation minus precipitation (i.e., dry locations will get dryer, wet locations moister) and its temporal variance. Changes in climate will also modify tropospheric chemistry and hence the concentrations of pollutants arriving at downwind continents. Increased water vapour in a future warmed atmosphere leading to increased ozone destruction and shorter ozone lifetimes is a prominent and robust multi-model feature [*Stevenson et al*., 2006]. This effect may cause a reduction in the contribution of Asian emissions to background ozone over the United States in a future climate [*Murazaki and Hess*, 2006]. On the other hand, a number of studies [*Murazaki and Hess*, 2006; *Hauglustaine et al*., 2005] have shown increased local production of ozone in high emission areas, leading to increased ozone export, at least locally. In addition, ICT of ozone may also be influenced by enhanced lightning NO_x emissions [*Hauglustaine et al*., 2005] and enhanced stratosphere-troposphere exchange (STE) in a warmer climate [*Collins et al*., 2003; *Zeng and Pyle*, 2003; *Stevenson et al*. 2006].

A number of General Circulation Model (GCM) studies, although not all, have reported a tendency towards fewer but more intense mid-latitude storms in the future [*Cubasch et al*., 2001; *Lambert and Fyfe*, 2005], a poleward shift of mid-latitude storms [*Yin*, 2005; *Bengtsson et al*., 2006] and an associated increase in cyclonic circulation patterns over the Arctic [*Cassano et al*,. 2005]. Chemistry transport model simulations performed under future SRES A1(b) climate scenarios also suggest a decrease in synoptic activity (intensity and frequency) over the USA [*Mickley et al.* 2004; *Murazaki and Hess*, 2006], in agreement with the GCM studies above. A decrease in synoptic activity would result in less long-range pollutant transport driven by synoptic systems [*Holzer and Boer*, 2001], but possibly an increased role for convection and high altitude transport.

There is also a positive trend in the North Atlantic Oscillation (NAO) projected by the majority of GCMs [*Miller et al*., 2006], associated with a NE shift in Atlantic storm track activity, which may allow the Arctic to become more polluted in the future [*Eckhardt et al*., 2003]. *Hess and Lamarque* [2007] show that a positive Arctic Oscillation (AO) results in increased transport of U.S. emissions to Europe. However, model projections vary widely in their magnitude of response [*Osborn*, 2004; *Miller et al*., 2006]. These results concur with observational evidence of a poleward shift of the storm track [*McCabe et al*., 2001] and a positive trend in the Northern Annular Mode (NAM) or AO, and NAO [*Harnik and Chang*, 2003] during the 1970s-1990s. *Liu et al*. [2005] relate stronger eastward transport of East Asian emissions to El Niño events; a future trend towards more El Niño-like conditions has been noted in a number of (but not all) studies [*Collins*, 2005].

2.5 Outstanding issues and recommendations

2.5.1 Basic transport mechanisms

Basic large-scale mechanisms for transporting pollutants out of the boundary layer have been reasonably well documented in both measurements and models, however the importance of small scale venting on intercontinental transport, such as dry convection, local circulations (valley, or land-ocean) and gravity wave mixing has not. These smaller scale venting mechanisms have greatest importance during summer due to diminished large-scale stirring by synoptic systems, especially for low-altitude transport events from Asia to the U.S. [*Holzer et al*. 2005]. These types of events are difficult to observe (i.e., they are not associated with large-scale plume transport) and are not often resolved in large-scale model simulations.

The transport of pollutants from the free troposphere to the surface is also not well observed, either in large-scale models or in observations. This may be partly due to the fact that descent into the boundary layer often does not involve large concentrated plumes, but background air. In addition, venting through the top of the boundary layer often involves small scale processes not directly simulated on the global scale.

The transport of species out of the boundary layer is often associated with wet-deposition as water vapour is condensed during the transport events. The extent to which soluble species are wet deposited depends on details of the microphysics i.e. to what extent: 1) convective clouds are able to transport soluble species to the upper troposphere, 2) soluble species are retained as liquid water freezes, and 3) rain is evaporated as it falls through the atmosphere. Again, these processes are not suitably modelled on the large scale and have not been adequately measured.

Finally transport in the tropics and subtropics has received less attention than in the mid-latitudes. With the growth of future emissions expected in the tropics and subtropics these processes will gain greater importance. Furthermore, the nature of the transport barrier between the tropics and mid-latitudes is not particularly well known.

2.5.2 Modelling

While large-scale transport is reasonably well represented in global models, parameterized processes are not: particularly deep convection, boundary layer mixing, boundary layer venting, and washout [*Lawrence and Rasch*, 2005; *Murazaki and Hess*, 2006]. For example, *Auvray et al.* [2007] find differences in the sign of net chemical tendencies in the mid-troposphere in the North Atlantic and North Pacific, which they attribute to differences in model transport schemes and water vapour transport as well as lightning. These processes are critical for venting pollutants into and out of the boundary layer. Uncertainties in the transport of soluble pollutants are high. Processes that are influenced by sub-grid-scale orography are also a key uncertainty for models; venting and mixing into the boundary layer by land-sea breezes, mountain-valley breezes, gravity waves and flow over mountains are not well represented or not represented at all. These issues may be partly addressed through coupling between regional and global models.

Although source-receptor relationships have been extensively studied at the continental and country levels, studies to quantify the impacts of emissions at fine spatial scales such as mega-cities [*Lawrence et al.*, 2006] are important given the rapid and future projected growth of world cities. Model uncertainty associated with the methodology used for estimating source-receptor relationships also deserves attention. *Derwent et al*. [2004] find significant differences in estimates of the magnitude of intercontinental transport of ozone using tracer labelling vs. emission sensitivity techniques associated with non-linearities in ozone chemistry. Since source-receptor relationships require an accurate knowledge of the emission source, more sophisticated estimate methodologies and a better quantification of model error are also necessary. The ongoing HTAP calculations should help quantify inter-model sensitivities to intercontinental transport. Finally, our knowledge of the interannual variability of intercontinental transport and the role of large-scale modes of climate variability such as the NAO and ENSO, needs to be better quantified through multi-decadal model studies [e.g. *Duncan and Bey*, 2004; *Hess and Lamarque*, 2007; *Liu et al*., 2003b; *Liu et al*., 2005].

2.5.3 Measurements networks

To fully understand the impact of upwind emissions on downwind continents, a comprehensive in situ measurement network must be established to monitor trace gases and particulate matter along the inflow and outflow boundaries of each continent throughout the entire tropospheric column and the lowermost stratosphere. No such network currently exists and the logistical and monetary barriers to its establishment are formidable. For the time being the most accessible observational information on pollutant import and export will have to come from remote sensing by polar orbiting instruments. The ENVISAT SCIAMACHY instrument provides column NO_2 measurements while the AIRS instrument on the NASA AQUA satellite provides column CO measurements, and the MISR instrument on TERRA measures aerosol optical depth. The NASA AURA satellite can monitor ozone, HNO_3, CH_4, aerosol extinction, CO, and SO_2 in the upper troposphere using the HIRDLS and MLS limb sounders, while the OMI instrument can provide column measurements of ozone, SO_2, HCHO, NO_2, aerosol optical thickness and aerosol single scattering albedo. Most importantly the TES instrument on AURA provides tropospheric profiles of O_3, CH_4 and CO, and upper tropospheric profiles of

HNO_3 and NO_2. While these instruments provide global coverage the frequency at which they pass over a location means the data are best suited for monthly or seasonal averages. Other limitations are their inability to make measurements within and below cloud (although MLS can measure some trace gases in the presence of tropical cirrus), the coarse vertical resolution of the nadir viewing instruments (with TES having the best resolution of 2 km), and the limitation of MLS to only view the tropical upper troposphere.

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