Chapter 2: Conceptual Overview of Hemispheric or Intercontinental Transport Processes

Alphabetical list of authors:
O. R. Cooper, R. Doherty, P. Grennfelt, P. Hess and A. Stohl

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. In terms of emissions we focus on nitrogen oxides (NO_x) because of their primary origin in fossil fuel combustion, and their contributions to acid rain, and ozone production. The great majority of humans (88%) and anthropogenic NO_x emissions (91%, year 2000 EDGAR estimate) [*Olivier and Berdowski*, 2001] are located in the northern hemisphere. The percentages of global human population and global NO_x emissions distributed across four primary regions of the northern hemisphere are as follows: North America, 8% population (22% NO_x); Europe, 13% (25%); Asia, 54% (33%); northern Africa and the Middle East, 13% (9%). The major NO_x emission regions of the northern hemisphere are located in eastern USA/southeastern Canada, western and central Europe, and southern and eastern Asia. The major emission 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. Furthermore, these two regions plus south Asia commonly experience deep convection during spring and summer which can quickly transport pollutants from the boundary layer to the upper troposphere. 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].

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 [e.g. **Derwent et al.* 2001, *Wild et al.* 2001/4, *Fiore et al.* 2002], as discussed in section 2.5.2. Source-receptor relationships can be interpreted as: 1) due to the direct transport of plumes from the source to the receptor site and 2) the contribution of the source to the overall background tropospheric concentrations at a receptor site. While the transport of plumes involves the consideration of a number of distinct episodic transport events occuring 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 (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. On the other hand, the source-receptor relationship of long-lived species (e.g., methane) will mostly be determined by the contribution of the source to the background component of the distribution.

Figure XXI (figure still being produced) shows the zonal mean tropospheric winds for January and July, demonstrating a number of salient points with regards to intercontinental transport. 1) Zonal mean winds in the mid-latitudes 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 pollutant plumes into the mid- to upper troposphere (e.g., convection or transport by synoptic systems) 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. While not shown, meridional winds are generally weaker than zonal winds, thus pollutants tend to be transported zonally.

Figure XX1 also shows lines of potential temperature. These strongly constrain transport, which follows isentropic surfaces in the absence of heating. Sensible heating at the Earth's surface and the latent heat of condensation are the primary processes by which the atmosphere is warmed; these processes are balanced in the mean by radiational cooling (1-2 degrees per day, on average). In the free troposphere the heating of air generally occurs on small spatial and temporal scales, due to the condensation of water vapor within convective updrafts or within the warm sector of mid-latitude cyclones, particularly the warm conveyor belt. On the other hand, subsidence and tropospheric cooling occur on large spatial scales and long timescales. The long timescale for radiational cooling implies a strong constraint on the potential temperature difference between surface source and receptor locations, severely limiting the long-range transport processes of tropospheric plumes. In particular, since potential temperatures generally decrease towards the poles, the poleward transport of pollution plumes is quite limited. In addition, processes which raise the potential temperature of air parcels above that of the earth's surface are not favored (e.g., deep convection; Hess, 2005). On the otherhand, the modest increases of potential temperature in mid-latitude warm conveyor belts (on the order of 15-20^o K; *Wernli et al. ????) can be an effective means for the long-range transport of pollutant plumes especially during the winter season.

A number of studies have shown semi-permeable transport barriers between the extra-tropics and the tropics [**Pierrehumbert and Yang*, 1993; **Bowman and Carrie*, 2002; **Bowman and Erukhimova*, 2004; *Hess*, 2005]. Thus a mid-latitude tracer source will most immediately affect the background tracer concentration within the mid-latitudes and only slowly affect tropical sites. 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 N.H. and S.H. tracer concentrations approaching 2 years [**Bowman and Erukhimova*, 2004]. Equatorward transport between the extratropics and tropics occurs primarily in shallow flow near the earth's surface [**Bowman and Carrie*, 2002; **Bowman and Erukhimova*, 2004]. This flow corresponds to the equatorward branch of the mean-diabatic circulation [*Held and Schneider*, 1999] and occurs at temperatures below that of the mean surface temperature (i.e., on the poleward side of coldfronts). Poleward flow occurs in a more diffuse circulation in the upper troposphere.

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 the 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 (FIG. X1). Due to the upward and poleward slope of the isentropic surfaces, poleward moving air tends to ascend, while equatorward moving air tends to sink. While dry convection, shallow convection, mountain valley circulations, 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.

The convectively influenced regime occurs predominantly within the tropics [*Hess*, 2005]. Here rapid ascending motion occurs predominantly 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. Meridional stirring occurs along isentropic surfaces oriented approximately parallel to the earth's surface on a timescale of approximately 20 days [**Bowman and Carrie*, 2002; **Bowman and Erukhimova*, 2004]. The N.H. summertime mid-latitude mid- and upper-troposphere is also predominantly convectively influenced, as convection is a dominant mechanism in venting the continental boundary layer.

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 systems are weaker [*Merrill and Moody*, 1996; *Cooper et al*., 2002; *Stohl et al*., 2002]. The cyclones are typically composed of four different airstreams that have been shown to 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 northward, 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].

In terms of rapid intercontinental pollutant transport the WCB is the most important airstream 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 North 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 not only results in slightly longer transport times, but in some instances two WCBs are required to transport emissions from Asia to North America [*Cooper et al*., 2004].

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 off shore.

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].

Convection is an important mechanism for vertically transporting air pollutants [*Dickerson et al*., 1987; *Lelieveld and Crutzen*, 1994]. 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.

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]. The highest altitude where smoke from boreal forest fires was observed in situ was 17 km, several kilometers above the tropopause and at potential temperatures greater than 380 K [*Jost et al*., 2004]. Remote sensing observations indicate that even higher injections are possible [*Fromm et al*., 2005].

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*, 1998]. 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 [*Guerova et al.* 2006; *Li et al.* 2002].

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 North 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 difficult to detect. Focusing first on transport from Asia to North America, modelling studies indicate that episodic long-range transport of CO occurs from Asia to the Northeast Pacific region throughout the year every 10, 15 and 30 days in the upper, middle and lower troposphere, respectively [*Liang et al*., 2004] and that 3-5 Asian plumes impact the boundary layer of the U.S. west coast in a typical February-May time period [*Yienger et al*., 2000]. Aircraft studies have detected very strong Asian plumes in the lower and mid-troposphere above the eastern Pacific [*Heald et al*., 2003; *Nowak et al*., 2004] and the US west coast [*Jaffe et al*., 1999; *Jaffe et al.*, 2003; *Cooper et al.*, 2004ab] with CO on occasion reaching 300 ppbv. Most of these free tropospheric observations, and all with CO in excess of 200 ppbv were associated with WCB export from Asia, while the other 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 pollution events are not always associated with elevated ozone mixing ratios, and the enhancements are difficult to detect at the surface. A modeling study by *Hudman et al*. [2004] indicates that when the ozone exceeds 80 ppbv at mountaintop sites in California the average contribution from Asian ozone is about 7 ppbv. As Asian pollution plumes continue travelling east, clear enhancements of ozone and aerosol concentrations can also be found over Europe [Stohl et al., 2006]. Asian pollution over Europe has furthermore been documented after taking the alternative shorter pathway involving westward transport with the monsoon circulation from India to Africa and the Mediterranean [*Lawrence et al*., 2003].

In terms of large scale North American export events influencing Europe, confirmation of their strong impact has only been made in the free troposphere above Europe [*Stohl and Trickl*, 1999], or at high mountaintop sites. For example, two North American plumes transported to Europe by WCBs were intercepted by a research aircraft in the lower to mid- free troposphere [*Stohl et al*., 2003a; *Huntrieser et al*., 2005] with CO mixing ratios reaching 170 ppbv. Only one of the plumes was observed to impact the surface, at two monitoring sites in the Alps in the 3-4 km altitude range. Regarding the impact of North American plumes on low altitude surface sites in Europe, *Derwent et al.* [1998] found 5 probable cases of North American emissions influencing Mace Head on the west coast of Ireland, but the pollutant concentrations were quite low. To date the only major North 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]. Typically, this pollution traverses the Arctic and reaches high-latitude North America [*Sharma et al.*, 2006]. While early Arctic explorers observed atmospheric haze and dirty deposits on the snow [*Garrett and Verzella, 2007*], the remote Arctic atmosphere was long believed to be extremely clean. However, in the 1950s, pilots flying over the North American Arctic observed widespread haze [*Mitchell*, 1957] every winter and early spring. In the 1970s scientists realized that the haze was air pollution transported from the mid-latitudes [*Barrie*, 1986]. This

Arctic Haze is a mixture of sulfate and particulate organic matter and, to a lesser extent, ammonium, nitrate, black carbon, and dust aerosols [*Quinn et al.*, 2007].

Although it is likely that a large fraction of the European pollution is transported to Asia, there is a lack of studies of this transport pathway. *Newell and Evans* [2000] estimated that some 25% of the air parcels arriving over Central Asia have crossed over Europe before and some 4% have originated in the European boundary layer. *Pochanart et al*. [2003] have shown that average ozone and CO concentrations at a station in East Siberia are enhanced in air masses transported from Europe.

2.3.2 Contribution to background pollution

While enhanced plumes of pollution from upwind continents have been observed in the free troposphere above the west coasts of North America and Europe these events occur episodically on time scales of 1-2 weeks. *Liang et al*. [2004] show episodic long-range transport events occur rather infrequently off the northwest coast of the U.S. They characterize strong transport events which result in a 20-40 ppbv rise in observed CO levels, or about 15 to 25% of the concentration of CO. Ozone enhancements are even less [Liang?, Jaegle?]. This suggests that the concentration of species with intermediate lifetimes is mostly comprised of the background concentration. A number of studies have suggested changes in the background levels of pollutants along the west coast of North America [*Jaffe et al*. 2003; *Parrish et al*., 2004] and Europe [*Derwent* ??]. *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. Thus 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 the concentrations of background ozone were greater.

How does transport affect the background concentration of species? 1) It sets the distribution of background concentrations. The background concentrations of surface emissions tend to be oriented approximately along isentropic surfaces in the mid-latitudes due to the rapid mixing [*Plumb and McEwan*, 1979; **Bowman and Carrie*, 2002; **Bowman and Erukhimova*, 2004; *Hess*, 2005] along these surfaces. The atmospheric circulation itself tends to concentrate the highest concentrations of constituents towards the poles [*Hess*, 2005]. 2) The existence of transport barriers between the mid-latitudes and the subtropics suggests that background concentrations in the mid-latitudes will primarily be affected by mid-latitude emissions, and those in the subtropics and tropics by subtropical and tropical emissions. 3) Transport from the mid-latitudes to the subtropics largely occurs near the Earth’s surface in the subtropical boundary layer. This prolonged boundary layer transport tends to set the initial conditions for mid-latitude air transported to the tropics. In particular, ozone concentrations are highly depleted in the subtropical boundary layer. On the other hand tropical emissions enter the mid-latitudes above the boundary layer.

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.

Aerosols modify climate forcing directly by enhancing the scattering and absorption of solar radiation, and indirectly by acting as cloud condensation nuclei (CCN) to produce brighter clouds that are less efficient at releasing precipitation. These in turn lead to large reductions in the amount of solar irradiance reaching Earth's surface, a corresponding increase in solar heating of the atmosphere, changes in the atmospheric temperature structure, suppression of rainfall, and less efficient removal of pollutants, and a weaker hydrological cycle [*Ramanathan et al*. 2001a]. Long-range transport of aerosols has recently 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. The INDOEX campaign revealed the influence of black carbon absorbing aerosols in these atmospheric brown clouds on observed regional climate [*Ramanathan et al.* 2001a,b]. Regionally, the absorbing haze 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, with consequences for tropical rainfall and the hydrologic cycle [*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, which leads to multiple reflection and scattering of light between the surface and the aerosol layers. Black carbon, which is responsible for most of the aerosol light absorption, is a minor but important component of the Arctic Haze and causes a 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]. Its efficacy, measured as the effectiveness in increasing the surface air temperature per unit forcing, is twice as large as that of CO_2, and it may be even more effective in melting snow and ice. Numerous studies have also examined ICT of desert dust from Asia and North Africa [**Liu et al.*, 2003]. The transport of Saharan dust has recently been linked to hurricane suppression [*Dunion and Velden*, 2004]. These authors suggest that when the Saharan Air Layer engulfs tropical waves, or pre-existing tropical cyclones, its dry air, temperature inversion, and strong vertical wind shear (associated with the mid-level easterly jet) can inhibit their ability to strengthen. *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.*].

-Sources of black carbon on snow should also be mentioned. (Hansen, Stohl and other?)

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 (section 2.2). Predictions of changes in regional climate are generally less robust than large scale changes [*Giorgi et al.*, 2001]. Examining experiments generated for the 4th assessment report **Held and Soden* [2006] found a number of changes robust across the models: a decrease in deep convective mass flux**,** 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* ??] 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 an enhanced Brewer-Dobson circulation and 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]. There is also a positive trend in the North Atlantic Oscillation (NAO) projected by the majority of GCMs [**Miller et al.*, 2006], which is associated with a NE shift in Atlantic storm track activity. Most notably, pollution transport into the Arctic from Europe correlates positively with the NAO [*Eckhardt et al.*, 2003] such that in the future the Arctic may become more polluted. **Hess and Lamarque* [paper in press, JGR, 2007] also show that a positive AO results in increased transport of U.S. emission to Europe. However, model projections vary considerably in their magnitude of response [*Osborn*, 2004; *Miller et al.*, 2005]. 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 Arctic Oscillation (AO) and North Atlantic Oscillation (NAO) [*Harnik and Chan*g, 2003] between the 1970s-1990s.

Chemistry transport model (CTM) simulations performed under future SRES A1(b) climate scenarios also suggest a decrease in synoptic activity (intensity and frequency) over the United States [*Mickley et al.* 2004; *Murazaki and Hess*, 2006], in agreement with the GCM studies above. *Murazaki and Hess* [2006] find these simulated circulation changes to be reflected in simulated CO and ozone variability. A decrease in synoptic activity would result in less long-range pollutant transport driven through synoptic systems, but possibly an increased role for convection and high altitude transport.

Future increases in precipitation across the tropical oceans and in some of the monsoon regions are notable but not consistent amongst multi-model simulations [*Cubasch et al*., 2001]. A trend towards more El Niño-like conditions in the tropical Pacific has been noted in a number of GCMs, but again there is no overall model consensus [*Collins*, 2005]. *Liu et al*. [2005] (section 2.5.2) relate enhanced convection and stronger eastward transport of East Asian emissions in the subtropical eastern Pacific to El Niño winters. Future precipitation changes over the convective regions of the United States in summer is of inconsistent sign between GCMs [*Giorgi et al*., 2001]. Furthermore, future changes in convection will be accompanied by changes in lightning NO_x emissions, and hence ozone production.

Moreover, since the altitude of convective outflow has important implications for transport speed and direction and the chemical lifetime of lofted species [*Schultz and Bey*, 2004], changes in convection height and the strength of deepest convection may have important implications for ICT and chemistry in the troposphere [*Stevenson et al.*, 2005]. In a near-future CTM simulation, *Stevenson et al*. [2005] generally found an increase in convective updraught strength at 150 hPa in the tropics and sub-tropics. Overall, the concentrations of pollutants arriving at downwind continents in the future will be determined by future emissions at source locations, changes in chemistry due to changes in climate as well as lightning NO_x and STE, and changes in ICT patterns and their transport processes.

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. While the impact of smaller scale venting mechanisms on ICT may not be important during winter due to the frequent stirring of the mid-latitude troposphere by synoptic systems, these venting mechanisms have greater importance during summer. **Holzer et al*. [2006] show that low-altitude transport events from Asia to the U.S. are more prevalent in summer due to the lack of large-scale stirring by synoptic systems. The importance of small scale venting on ITC, such as dry convection, local circulations (ocean-valley, or land-ocean) and gravity wave mixing has not been well documented [but see…..]. 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: to what extent are convective clouds able to transport soluble species to the upper troposphere, to what extent are soluble species retained as liquid water freezes [**Barth et al…*], to what extent is rain 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

CTMs have been applied to investigate a number of issues relating to ICT namely: transport pathways and continental outflow and their seasonal variability, chemical tendencies in pollution plumes, source-receptor relationships and the potential impact of emission reductions from one continent on the pollutant concentration over downwind continents. A number of studies have recently investigated meteorological-driven interannual variability in ICT, associated with large-scale modes of variability such as the NAO and ENSO. Pollution export from North America to Europe [*Li et al.*, 2002] and from Europe to the Arctic [*Eckhardt et al*., 2003; *Duncan and Bey*, 2004; *Lamarque and Hess*, 2004,] has been correlated with the phase of the NAO/AO (see also section 2.4). Export of emissions from South East Asia to North America has been shown to be modulated by ENSO [**Liu et al.*, 2003; *Liu et al.*, 2005]. However, our knowledge of the interannual variability of ICT, needs to be better quantified through multi-decadal model studies.

The issue of source–receptor (S-R) relationships and the contribution of an upwind continent to concentrations of pollutants over downwind continents has also received much attention, including the studies mentioned above. These S-R relationship studies have generally been concerned with ICT of ozone and its precursors, although several recent studies have quantified the impact of Asian mineral dust aerosol emissions on visibility over the United States [*Heald et al*., 2006; *Park et al*., 2006]. However, all these studies have typically been performed for different time periods, using a range of methodologies and a single CTM. *Derwent et al*. [2004] note that tracer labelling and emission sensitivity techniques appear to give different estimates of the magnitude of ICT of ozone because of important non-linearities in the fast chemistry of ozone and its precursors. In addition, theses analyses have generally focussed on the continental or country scale. Most recently, *Lawrence et al*. [2006] examined ICT across 20 mega-cities in the tropics and mid-latitudes. They found the potential for pollution export from locations in the tropics to be greatest due to deep and frequent convection, and those affected by the monsoon exhibited the largest seasonality in their export potential. Source-receptor relationships require an accurate knowledge of the source. The source is often determined from bottom-up calculations. However, models also play a role through top-down calculations [**Petron, *Arrelano, *Kasibatla*]. However, **Arellano and Hess* have recently shown the top-down estimates are very dependent on the analyzed meteorology used. Thus more sophisticated estimate methodologies and a better quantification of model error is necessary. In general with all studies, the use of a single CTM precludes the quantification of uncertainty estimates associated with ICT.

While large-scale transport is reasonably well represented in global models, as discussed in section 2.5.1, 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.* [2006] 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 is high. Processes that are influenced by sub-grid-scale orography are also a key uncertainty for CTMs. Also mentioned in section 2.5.1, venting and mixing into the boundary layer by land-sea breezes, mountain-valley breezes, gravity waves and flow over mountains is not well represented or not represented at all. For example land-sea breezes might determine if pollutants are vented above of the boundary layer or transported within the boundary layer as pollutants flow over the water – this is critical in determining their losses. The points above may be partly addressed through coupling between regional and global models. The ongoing HTAP calculations should help quantify the model-model sensitivities to transport.

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 example, the monitoring of ozone, ozone precursors and particulate matter across the northern hemisphere would require a small fleet of highly instrumented aircraft dedicated to the daily monitoring along the five inflow and outflow boundaries of North America, Asia and Europe (the western and eastern edges of North America, the western edge of Europe, the eastern edge of Asia and down the spine of the Ural Mountains). The acquisition alone of 20 long-range aircraft (4 at each site: 2 high altitude and 2 low altitude) could require more than $1 billion USD if all aircraft were purchased new. In addition the annual operating costs to maintain the 20 aircraft and 5 support facilities would exceed $50 million USD. In comparison the estimated annual cost of leasing and maintaining a fleet of 36 unmanned aerial vehicles from 12 bases to collect daily global profiles of temperature, humidity and winds using dropsondes is on the order of $600 million USD, comparable to a satellite program such as the U.S. operational polar-orbiting satellite constellation [*Macdonald*, 2005].

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 AURA 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) and their coarse vertical resolution, with TES having the best resolution of 2 km.

References

References above marked with * indicate missing or uncertain full references below.

References need to be found for the following:
Derwent et al., 2001
Wernli et al.
Pierrehumbert and Yand, 1993
Bowman and Carrie, 2002
Bowman and Erukhimara, 2004
Held and Schneider, 1999
Plumb and McEwan, 1979
Warren and Wiscombe, 1980
Shindell et al.
Petron
Arrelano
Kasibatla
Arrelano and Hess

Angevine, W. M., M. P. Buhr, J. S. Holloway, M. Trainer, D. D. Parrish, J. I. MacPherson, G. L. Kok, R. D. Schillawski, and D. H. Bowlby (1996), Local meteorological features affecting chemical measurements at a North Atlantic coastal site, *J. Geophys. Res.*, 101(D22), 28,935–28,946.

Auvray, M., I. Bey, E. Llull, M.G. Schultz, and S. Rast, A model investigation of tropospheric ozone chemical tendencies in long-range transported pollution plumes, in press, J. Geophys. Res., 2006

Bader, M. J., G. S. Forbes, J. R. Grant, R. B. E. Lilley and A. J. Waters (Eds.), Images in weather forecasting: A practical guide for interpreting satellite and radar imagery, University Press, Cambridge, 1995.

Barrie, L. A. (1986), Arctic air pollution—An overview of current knowledge, *Atmos. Environ., 20*, 643– 663.

Bengtsson, L., K. I. Hodges and E. Roeckner, J., Storm Tracks and Climate Change, 2006, Clim., V19, 3518-3543.

Bethan, S., G. Vaughan, C. Gerbig, A. Volz-Thomas, H. Richer and D. A. Tiddeman, Chemical air mass differences near fronts, *J. Geophys. Res., 103*, 13,413-13,434, 1998.

Browning, K. A., and G. A. Monk, A simple model for the synoptic analysis of cold fronts, *Quart. J. R. Met. Soc., 108*, 435-452, 1982.

Browning, K. A., and N. M. Roberts, Structure of a frontal cyclone, *Q. J. R. Meteorol. Soc. 120*, 1537-1557, 1994.

Carlson, T. N., *Midlatitude Weather Systems*, Am. Meteorol. Soc., Boston, 1998.

Cassano, J.J., P. Uotila, and A. Lynch, 2005: Changes in synoptic weather patterns in the polar regions in the 20th and 21st centuries, Part 1: Arctic. Int. J. Clim., 26, 1181-1199, 2006

Collins W. J., R. G. Derwent, B. Garnier, C. E. Johnson, M. G. Sanderson, D. S. Stevenson, Effect of stratosphere-troposphere exchange on the future tropospheric ozone trend, J. Geophys. Res., 108 (D12), 8528, doi:10.1029/2002JD002617, 2003.

Cooper, O. R., J. L. Moody, D. D. Parrish, M. Trainer, J. S. Holloway, T. B. Ryerson, G. Hübler, F. C. Fehsenfeld, S. J. Oltmans and M. J. Evans, Trace gas signatures of the airstreams within North Atlantic cyclones - Case studies from the NARE'97 aircraft intensive, *J. Geophys. Res., 106*, 5437-5456, 2001.

Cooper, O. R., J. L. Moody, D. D. Parrish, M. Trainer, T. B. Ryerson, J. S. Holloway, G. Hübler, F. C. Fehsenfeld, and M. J. Evans, Trace gas composition of mid-latitude cyclones over the western North Atlantic Ocean: A conceptual model, *J. Geophys. Res.*, 107, 10.1029/2001JD000901, 2002a.

Cooper, O. R., C. Forster, D. Parrish, M. Trainer, E. Dunlea, T. B. Ryerson, G. Hübler, F. Fehsenfeld, D. Nicks, J. Holloway, J. Nowak, C. Brock, J. de Gouw, C. Warneke, J. Roberts, F. Flocke J. Moody, A case study of trans-Pacific warm conveyor belt transport: The influence of merging airstreams on trace gas import to North America, *J. Geophys. Res., 109*, D23S08, doi:10.1029/2003JD003624, 2004a.

Cooper, O. R., et al. (2004b), On the life-cycle of a stratospheric intrusion and its dispersion into polluted warm conveyor belts, *J. Geophys. Res., 109*, D23S09, doi:10.1029/2003JD004006.

Cotton, W. R., G. D. Alexander, R. Hertenstein, R. L. Walko, R. L. McAnelly, M. Nicholls (1995), Cloud venting - a review and some new global annual estimates, *Earth-Science Reviews*, 39, 169-206.

Cubasch, U., Meehl, G. A., Boer, G. J., Stouffer, R. J., Dix, M., Noda, A., Senior, C. A., Raper, S. and Yap, K. S., Projections of future climate change, In: Climate Change 2001: The Scientific Basis, Contribution of WG1 to the Third Assessment report of the IPCC, Eds. Houghton, J.T., et al., Cambridge University Press, England, 2001.

Derwent, R. G., P. G. Simmonds, S. Seuring and C. Dimmer, Observation and interpretation of the seasonal cycles in the surface concentrations of ozone and carbon monoxide at Mace Head, Ireland from 1990 to 1994, *Atmos. Environ.*, *32*, 145-157, 1998.

Derwent R. G., D. S. Stevenson, W. J. Collins, C. E. Johnson, Intercontinental transport and the origins of ozone observed at surface sites in Europe, Atmos. Environ., 38, 1891-1901, 2004

Dickerson, R. R., et al. (1987), Thunderstorms: An important mechanism in the transport of air pollutants. *Science*, 235**,** 460–465.

Duncan, B.N., and I. Bey, A modeling study of export pathways of pollution from Europe: Seasonal and Interannual variations (1987-1997), J. Geophys. Res., 109, D08301, doi:10.1029/2003JD004079, 2004

Dunion, J. P., and C. S. Velden (2004), The impact of the Saharan air layer on Atlantic tropical cyclone activity, Bull. Am. Meteorol. Soc., 85(3), 353–365. 2002JD002670.

Eckhardt, S. , Stohl, A., Beirle, S., Spichtinger, N., James, P., Forster, C., Junker, C., Wagner, T., Platt, U. and Jennings, S. G., The North Atlantic Oscillation controls air pollution transport to the Arctic, Atmospheric Chemistry and Physics, Vol. 3, pp 1769-1778, 2003

Eckhardt, S., A. Stohl, H. Wernli, P. James, C. Forster, and N. Spichtinger (2004): A 15-year climatology of warm conveyor belts, *J. Climate 17*, 218-237.

Evan A. T., J. Dunion, J. A. Foley, A. K. Heidinger, C. S. Velden (2006), New evidence for a relationship between Atlantic tropical cyclone activity and African dust outbreaks, Geophys. Res. Lett., 33, L19813, doi:10.1029/2006GL026408.

Fiore, A.M., D.J. Jacob, I. Bey, R.M. Yantosca, B.D. Field, A.C. Fusco, and J.G. Wilkinson, Background ozone over the United States in summer: origin,trend, and contribution to pollution episodes, J. Geophys. Res., 107 (D15), doi:10.1029/2001JD000982, 2002.

Forster, C., U. Wandinger, G. Wotawa, P. James, I. Mattis, D. Althausen, P. Simmonds, S. O'Doherty, S. G. Jennings, C. Kleefeld, J. Schneider, T. Trickl, S. Kreipl, H. Jäger, A. Stohl, Transport of boreal forest fire emissions from Canada to Europe, J. Geophys. Res., 106(D19), 22887-22906, 10.1029/2001JD900115, 2001.

Fromm, M., J. Alfred, K. Hoppel, J. Hornstein, R. Bevilacqua, E. Shettle, R. Servranckx, Z. Li, and B. Stocks (2000), Observations of boreal forest fire smoke in the stratosphere by POAM III, SAGE II, and lidar in 1998, *Geophys. Res. Lett.*, **27**, 1407–1410.

Fromm M, Bevilacqua R, Servranckx R, et al. (2005), Pyro-cumulonimbus injection of smoke to the stratosphere: Observations and impact of a super blowup in northwestern Canada on 3-4 August 1998, *J. Geophys. Res.*, 110, D08205, doi:10.1029/2004JD005350.

Garrett, T.J., and L. L. Verzella, *Bull. Am. Met. Soc.*, submitted (2007).

Giorgi, F., et al., Regional climate Information- Evaluations and Projections, In: Climate Change 2001: The Scientific Basis, Contribution of WG1 to the Third Assessment report of the IPCC, Eds. Houghton, J.T., et al., Cambridge University Press, England, 2001.

Guerova, G., I. Bey, J.-L. Attié, R. V. Martin, J. Cui, and M. Sprenger, Impact of transatlantic transport episodes on summertime ozone in Europe, Atmos. Chem. Phys., 6, 2057-2072, 2006

Hansen, J., and L. Nazarenko (2004), Soot climate forcing via snow and ice albedos, Proc. Natl. Acad. Sci. U.S.A., 101, 423 – 428, doi:10.1073/pnas.2237157100.

Harnik N. and E.K.M. Chang, 2003: Storm track variations as seen in radiosonde observations and reanalysis data. *J. Climate*, 16, 480–495.

Heald, C. L., et al. (2003), Asian outflow and trans-Pacific transport of carbon monoxide and ozone pollution: An integrated satellite, aircraft, and model perspective, *J. Geophys. Res.*, 108(D24), 4804, doi:10.1029/2003JD003507.

Heald, C. L., D. J. Jacob, R. J. Park, B. Alexander, T. D. Fairlie, R. M. Yantosca, and D. A. Chu (2006), Transpacific transport of Asian anthropogenic aerosols and its impact on surface air quality in the United States, *J. Geophys. Res.*, *111*, D14310, doi:10.1029/2005JD006847.

Henne, S., M. Furger, S. Nyeki, M. Steinbacher, B. Neininger, S. F. J. deWekker, J. Dommen, N. Spichtinger, A. Stohl, and A. S. H. Prévôt (2004): Quantification of topographic venting of boundary layer air to the free troposphere. *Atmos. Chem. Phys.* 4, 497-509.

Hess, P. G. (2005), A comparison of two paradigms: The relative global roles of moist convective versus nonconvective transport, J. Geophys. Res., 110, D20302, doi:10.1029/2004JD005456.

Hudman, R. C., D. J. Jacob, O.R. Cooper, M. J. Evans, C. L. Heald, R. J. Park, F. Fehsenfeld, F. Flocke, J. Holloway, G. Hübler, K. Kita, M. Koike, Y. Kondo, A. Neuman, J. Nowak, S. Oltmans, D. Parrish, J. M. Roberts, and T. Ryerson, Ozone production in transpacific Asian pollution plumes and implications for ozone air quality in California, *J. Geophys. Res.*, *109*, D23S10, doi:10.1029/2004JD004974, 2004.

Huntrieser, H., et al. (2005), Intercontinental air pollution transport from North America to Europe: Experimental evidence from airborne measurements and surface observations, *J. Geophys. Res.*, 110, D01305, doi:10.1029/2004JD005045.

Jaegle´, L., D. A. Jaffe, H. U. Price, P. Weiss-Penzias, P. I. Palmer, M. J. Evans, D. J. Jacob, and I. Bey (2003), Sources and budgets for CO and O3 in the northeastern Pacific during the spring of 2001: Results from the PHOBEA-II Experiment, J. Geophys. Res., 108(D20), 8802, doi:10.1029/2002JD003121.

Jaffe, D., et al. (1999), Transport of Asian air pollution to North America, Geophys. Res. Lett., 26, 711– 714.

Jaffe, D., I. McKendry, T. Anderson, and H. Price (2003a), Six 'new' episodes of trans-Pacific transport of air pollutants, *Atmos. Environ.*, *37*, 391– 404.

Jaffe, D., H. Price, D. D. Parrish, A. Goldstein, and J. Harris (2003b), Increasing background ozone during spring on the west coast of North America, *Geophys. Res. Lett.*, *30*(12), 1613, doi:10.1029/2003GL017024.

Jost, H.-J., K. Drdla, A. Stohl, L. Pfister, M. Loewenstein, J. P. Lopez, P. K. Hudson, D. M. Murphy, D. J. Cziczo, M. Fromm, T. P. Bui, J. Dean-Day, C. Gerbig, M. J. Mahoney, E. C. Richard, N. Spichtinger, J. V. Pittman, E. M. Weinstock, J. C. Wilson, and I. Xueref (2004), In-situ observations of midlatitude forest fire plumes deep in the stratosphere. *Geophys. Res. Lett.* 31, L11101, doi:10.1029/2003GL019253.

Kiley, C. M., and H. E. Fuelberg (2006), An examination of summertime cyclone transport processes during Intercontinental Chemical Transport Experiment (INTEX-A), *J. Geophys. Res.*, 111, D24S06, doi:10.1029/2006JD007115.

Klonecki, A., P. Hess, L. Emmons, L. Smith, J. Orlando, and D. Blake (2003), Seasonal changes in the transport of pollutants into the Arctic troposphere -- model study, J. Geophys. Res., 108, 8367, doi:10.1029/2002JD002199.

Lamarque J-F., P. G. Hess (2004), Arctic Oscillation modulation of the Northern Hemisphere spring tropospheric ozone, Geophys. Res. Lett., 31, L06127, doi:10.1029/2003GL019116

Lambert, S.J., and J.C. Fyfe, 2005: Changes in winter cyclone frequencies and strengths simulated in enhanced greenhouse warming experiments: results from the models participating in the IPCC 4

Lawrence, M. G., and P. J. Rasch (2005): Tracer transport in deep convective updrafts: Plume ensemble versus bulk formulations**,** *J. Atmos. Sci.*, **62**, 2880-2894.

Lawrence, M. G., Rasch, P. J., von Kuhlmann, R., Williams, J., Fischer, H., de Reus, M., Lelieveld, J., Crutzen, P. J., Schultz, M., Stier, P., Huntrieser, H., Heland, J., Stohl, A., Forster, C., Elbern, H., Jakobs, H., and Dickerson, R. R.: Global chemical weather forecasts for field campaign planning: predictions and observations of large-scale features during MINOS, CONTRACE and INDOEX. Atmos. Chem. Phys. 3, 267-289, 2003.

Lawrence, M.G., T. M. Butler, J. Steinkamp, B. R. Gurjar, J. Lelieveld ,Regional pollution potentials of megacities and other major population centers, atmos.Chem.Phys. disc. 6, 13323-13366, 2006

Lelieveld, J., and P. J. Crutzen (1994): Role of deep cloud convection in the ozone budget of the troposphere. *Science*, 264**,** 1759–1761.

Li Q., D. J. Jacob, I. Bey, P. I. Palmer, B. N. Duncan, B. D. Field, R. V. Martin, A. M. Fiore, R. M. Yantosca, D. D. Parrish, P. G. Simmonds, and S. J. Oltmans, Transatlantic transport of pollution and its effects on surface ozone in Europe and North America, J. Geophys. Res., 107 (D13), doi:10.1029/2001JD001422, 2002.

Liang, Q., L. Jaegle´, D. A. Jaffe, P. Weiss-Penzias, A. Heckman, and J. A. Snow (2004), Long-range transport of Asian pollution to the northeast Pacific: Seasonal variations and transport pathways of carbon monoxide, *J. Geophys. Res., 109*, D23S07, doi:10.1029/2003JD004402.

Liu, H., D.J. Jacob, I. Bey , R.M. Yantosca, B.N. Duncan, and G.W. Sachse, Transport pathways for Asian combustion outflow over the Pacific: Interannual and seasonal variations, J. Geophys. Res., 108(D20), 8786, doi: 10.1029/2002JD003102, 2003a.

Liu M., D. L. Westphal, S. Wang, A. Shimizu, N. Sugimoto, J. Zhou, Y. Chen, A high-resolution numerical study of the Asian dust storms of April 2001, J. Geophys. Res., 108 (D23), 8653, doi:10.1029/2002JD003178, 2003b.

Liu, J., Mauzerall, D. L., Horowitz, L.W., “Analysis of Seasonal and Interannual Variability in Transpacific Transport,” J. Geophys. Res., 110, D04302, doi: 10.1029/2004JD005207, 2005.

Macdonald, A. E., A global profiling system for improved weather and climate prediction, *Bull. Amer. Met. Soc*., 86, 1747-1764, 2005.

McCabe, G.J., M.P. Clark, and M.C. Serreze, 2001: Trends in Northern Hemisphere surface cyclone frequency and intensity. *J. Climate*, 14, 2763–2768.

Merrill, J. T., and J. L. Moody, Synoptic meteorology and transport during the North Atlantic Regional Experiment (NARE) intensive: Overview, *J. Geophys. Res., 101*, 28,903-28,921, 1996.

Mickley L. J., D. J. Jacob, B. D. Field, D. Rind (2004), Effects of future climate change on regional air pollution episodes in the United States, Geophys. Res. Lett., 31, L24103, doi:10.1029/2004GL021216.

Miller, R.L., G.A. Schmidt, and D.T. Shindell, 2005: Forced variations of annular modes in the 20th century IPCC AR4 simulations. J. Geophys. Res., submitted. 36

Mitchell, J. M., 1957: Visual range in the polar regions with particular reference to the Alaskan Arctic, J. Atmos. Terr. Phys. Special Suppl., 195-211.

Murazaki K., P. Hess (2006), How does climate change contribute to surface ozone change over the United States?, J. Geophys. Res., 111, D05301, doi:10.1029/2005JD005873.

Neuman, J. A., D. D. Parrish, M. Trainer, T. B. Ryerson, J. S. Holloway, J. B. Nowak, A. Swanson, F. Flocke, J. M. Roberts, S. S. Brown, H. Stark, R. Sommariva, A. Stohl, R. Peltiers, R. Weber, A. Wollny, D. T. Sueper, G. Hubler, and F. C. Fehsenfeld (2006): Reactive nitrogen transport and photochemistry in urban plumes over the North Atlantic Ocean. *J. Geophys. Res.* 111, D23S54, doi:10.1029/2005JD007010.

Newell, R. E., and M. J. Evans, Seasonal changes in pollutant transport to the North Pacific: The relative importance of Asian and European sources, Geophys. Res. Lett. 27, 2509-2512, 2000.

Nowak, J. B., et al. (2004), Gas-phase chemical characteristics of Asian emission plumes observed during ITCT 2K2 over the eastern North Pacific Ocean, *J. Geophys. Res.*, 109, D23S19, doi:10.1029/2003JD004488.

Olivier, J. G. J., and J. J. M. Berdowski (2001), Global emissions sources and sinks, in The Climate System, edited by J. Berdowski, R. Guicherit, and B. J. Heij, pp. 33–78, A. A. Balkema, Brookfield, Vt.

Osborn, T.J., 2004: Simulating the winter North Atlantic Oscillation: The roles of internal variability and greenhouse forcing. Clim. Dyn., 22, 605-623. 42

Owen, R. C., O. R. Cooper, A. Stohl, and R. E. Honrath (2006): An analysis of the mechanisms of North American pollutant transport to the Central North Atlantic lower free troposphere. *J. Geophys. Res.* 111, D23S58, doi:10.1029/2006JD007062.

Park, R. J., D. J. Jacob, N. Kumar, and R. M. Yantosca (2006), *"Regional visibility statistics in the United States: Natural and transboundary pollution influences, and implications for the Regional Haze Rule"*, Atmos. Environ., 40(28), 5405-5423.

Parrish, D. D., et al. (2004), Changes in the photochemical environment of the temperate North Pacific troposphere in response to increased Asian emissions, J. Geophys. Res., 109, D23S18, doi:10.1029/2004JD004978.

Pochanart, P., H. Akimoto, Y. Kajii, V. M. Potemkin, T. V. Khodzher (2003): Regional background ozone and carbon monoxide variations in remote Siberia/East Asia, J. Geophys. Res. 108, 4028.

Quinn, P. K., G. Shaw, E. Andrews, E. G. Dutton, T. Ruoho-Airola, and S. L. Gong (2007): Arctic haze: current trends and knowledge gaps, *Tellus*, in press.

Ramana M. V., V. Ramanathan (2006), Abrupt transition from natural to anthropogenic aerosol radiative forcing: Observations at the ABC-Maldives Climate Observatory, J. Geophys. Res., 111, D20207, doi:10.1029/2006JD007063.

Ramanathan, V., et al., 2001a: Aerosols, climate and the hydrological cycle. *Science*, 294, 2119–2124.

Ramanathan, V., et al., 2001b, Indian Ocean experiment: An integrated analysis of the climate forcing and effects of the great Indo-Asian haze, J. Geophys. Res., 106(D22), 28371-28398, 10.1029/2001JD900133, 2001.

Schultz and Bey, 2004: Numerical Modelling of Long-range pollution transport in Intercontinental transport of Air Pollution, ed. A. Stohl, chapter 10, 197-223, Springer, Berlin.

Sharma, S., E. Andrews, L. A. Barrie, J. A. Ogren, and D. Lavoue´ (2006): Variations and sources of the equivalent black carbon in the high Arctic revealed by long-term observations at Alert and Barrow: 1989-2003, J. Geophys. Res. 111, D14208, doi:10.1029/2005JD006581.

Speidel, M., R. Nau, F. Arnold, H. Schlager, and A. Stohl (2007), Aircraft-based atmospheric sulfur dioxide measurements during ITOP 2004: Evidence for transatlantic transport. In preparation for *J. Geophys. Res*.

Stevenson, D.S., R.M. Doherty, M.G. Sanderson, C.E. Johnson, W.J.Collins, and R.G. Derwent, Impacts of climate change and variability on tropospheric chemistry, Faraday Discussions, DOI: 10.1039/b417412g, 2005.

Stevenson D. S., et al. (2006), Multimodel ensemble simulations of present-day and near-future tropospheric ozone, J. Geophys. Res., 111, D08301, doi:10.1029/2005JD006338.

Stohl, A., and T. Trickl, A textbook example of long-range transport: Simultaneous observation of ozone maxima of stratospheric and North American origin in the free troposphere over Europe, J. Geophys. Res. 104, 30,445-30,462, 1999.

Stohl, A., S. Eckhardt, C. Forster, P. James, and N. Spichtinger, On the transport and timescales of intercontinental air pollution transport, *J. Geophys. Res.*, *107*(D23), 4684, doi:10.1029/2001JD001396, 2002.

Stohl, A., C. Forster, S. Eckhardt, N. Spichtinger, H. Huntrieser, J. Heland, H. Schlager, H. Aufmhoff, F. Arnold and O. Cooper, A backward modeling study of intercontinental pollution transport using aircraft measurements, *J. Geophys. Res.*, *in-press*, 2003a.

Stohl, A., H. Huntrieser, A. Richter, S. Beirle, O. Cooper, S. Eckhardt, C. Forster, P. James, N. Spichtinger, T. Wagner, M. Wenig, J. Burrows, and U. Platt, Rapid intercontinental air pollution transport associated with a meteorological bomb, *Atmos. Chem. Phys.*, *3*, 969-985, 2003b.

Stohl, A. (2006): Characteristics of atmospheric transport into the Arctic troposphere. *J. Geophys. Res.* 111, D11306, doi:10.1029/2005JD006888.

Stohl, A., C. Forster, H. Huntrieser, H. Mannstein, W. W. McMillan, A. Petzold, H. Schlager, and B. Weinzierl (2006): Aircraft measurements over Europe of an air pollution plume from Southeast Asia – aerosol and chemical characterization. *Atmos. Chem. Phys. Discuss.* **6**, 12611–12670.

Stull, R. B.: An Introduction to Boundary Layer Meteorology. Kluwer Academic Publishers, Dordrecht, 1988.

Wild, O., H. Akimoto, Intercontinental transport of ozone and its precursors in a three-dimensional global CTM, J. Geophys. Res., 106(D21), 27729-27744, 10.1029/2000JD000123, 2001.

Wild O., P. Pochanart, H. Akimoto (2004), Trans-Eurasian transport of ozone and its precursors, *J. Geophys. Res.*, *109*, D11302, doi:10.1029/2003JD004501.

Wild, M., et al., 2004: On the consistency of trends in radiation and temperature records and implications for the global hydrological cycle. *Geophys. Res. Lett.*, 31, L11201, doi:10.1029/2003GL019188.

Yienger, J. J., M. Galanter, T. A. Holloway, M. J. Phadnis, S. K. Guttikunda, G. R. Carmichael,W. J.Moxim, and H. Levy II (2000), The episodic nature of air pollution transport from Asia to North America, *J. Geophys. Res.*, *105*, 26,931– 26,945.

Yin, J.H., 2005: A consistent poleward shift of the storm tracks in simulations of 21st century climate. Geophys. Res. Lett., submitted. 52

Zeng G., and J. A. Pyle, Changes in tropospheric ozone between 2000 and 2100 modeled in a chemistry-climate model, Geophys. Res. Lett., 30 (7), 1392, doi:10.1029/2002GL016708, 2003.