

Review

# Intercontinental transport of aerosols and photochemical oxidants from Asia and its consequences

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*Air quality over parts of North America is being affected by pollutants transported from Asia.*

## Abstract

The intercontinental transport of aerosols and photochemical oxidants from Asia is a crucial issue for air quality concerns in countries downwind of the significant emissions and concentrations of pollutants occurring in this important region of the world. Since the lifetimes of some important pollutants are long enough to be transported over long distance in the troposphere, regional control strategies for air pollution in downwind countries might be ineffective without considering the effects of long-range transport of pollutants from Asia. Field campaigns provide strong evidence for the intercontinental transport of Asian pollutants. They, together with ground-based observations and model simulations, show that the air quality over parts of North America is being affected by the pollutants transported from Asia. This paper examines the current understanding of the intercontinental transport of gases and aerosols from Asia and resulting effects on air quality, and on the regional and global climate system.

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## 1. Introduction

Rapid industrialization in Asian countries in recent decades has resulted in a dramatic increase in the emissions of various air pollutants, including nitrogen oxides ( $\text{NO}_x$ ), sulfur dioxide ( $\text{SO}_2$ ) and carbon monoxide (CO) (e.g. Elliot et al., 1997; Van Aardenne et al., 1999). From the mid-1970s to the 2000s, emissions of both  $\text{NO}_x$  and  $\text{SO}_2$  over Asia increased at an average rate of  $\sim 4\%$ /year. In future decades, with an assumed continuation of significant economic growth, further increases in Asian anthropogenic emissions are expected. In addition, as an important natural pollutant, mineral dust is emitted into the atmosphere from arid regions in Asia almost every year, with its long-range transport often observed in satellite measurements.

The increasing anthropogenic emissions and persistent dust emissions not only have reduced the air quality in Asia itself, but also are affecting the pollutant deposition into the Pacific Ocean and air quality in downwind areas, including other countries in Asia, various islands in the Pacific region, and across the Pacific to North America. Previously studies have shown that gases and aerosols from Asia, including those secondary pollutants from tropospheric photochemistry, may affect the air quality and climate over downwind regions (e.g., Parrish et al., 1992; Heald et al., 2003, 2006; Nowak et al., 2004). Gaseous and aerosol pollutants, once deposited on the Pacific, may impact marine ecosystems. Various pollutants, including mineral dust, from Asia can be transported across the Pacific Ocean to North America, with resulting important consequences on local air quality (e.g., Prospero, 1999; Wei et al., 2002; Heald et al., 2006). On a longer time scale, the transported pollutants can affect both regional and global climate through their impacts on the atmospheric compositions and

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physical (e.g., acting as cloud condensation nuclei) and chemical (e.g., heterogeneous reactions) processes. Therefore the intercontinental transport of Asian photochemical oxidants and aerosols are a very important issue for air quality and climate concerns.

The effects of Asian pollutants on downwind areas have been of increasing interest during the last few decades. A variety of measurement and modeling studies during the past two decades have documented the intercontinental chemical transport of gases, mineral dust and other aerosols originated from Asia, including the secondary pollutants produced from atmospheric photochemistry. Earlier studies documented the long-range transport of dust (Prospero, 1979; Duce et al., 1980). More recently, several international research programs developed by various agencies from the U.S. and other countries have systematically studied the intercontinental transport of Asian pollutants and its possible impact on the air quality over downwind regions. Particularly relevant are those measurement campaigns developed under the auspices of the International Global Atmospheric Chemistry Project (IGAC): the Intercontinental Transport and Chemical Transformation (ITCT). The field campaigns carried out under ITCT include the Pacific Exploratory Mission-Western Pacific (PEM-West), the Asian Aerosol Characterization Experiments (ACE-Asia), the Transport And Chemical Evolution Over The Pacific (TRACE-P), and the Pacific Exploration of Asian Continental Emission (PEACE) (ITCT White Paper, 2005). These campaigns focus on the influence of Asian anthropogenic emissions on downwind concentrations of ozone and aerosols, through sampling and comparing the pollutant compositions along the Pacific coasts of Asia and North America together with accompanied model simulations (Jaffe et al., 2001; Jordan et al., 2003; Maxwell-Meier et al., 2004). Numerical modeling studies of intercontinental pollutant transport from Asia have examined the transport of ozone, sulfur compounds, nitrogen compounds, dust, soot, and persistent organic pollutants (POPs), focusing on various topics including emission estimations, transport pathways, and impacts on downwind air quality (e.g., Jacob et al., 1999; Wei et al., 2002; Hudman et al., 2004; Heald et al., 2006).

While past research has provided extensive insights into various issues related to Asian pollutant transport, many questions have yet to be answered. Major issues include accurate pollution observations and reliable emission estimations over Asian countries; better understanding of the pathways and transformations of the pollutants and their interactions with clouds in the transport processes; long-term measurements and analyses of the intercontinental transport; impacts of Asian pollutants on the marine ecosystems of the Pacific Ocean and the air quality and climate over North America. In particular, as China and India are increasing their energy and transportation infrastructures, pollutant emissions are also expected to increase in future years, but the resulting impacts from long-range transport processes remain highly uncertain.

This paper reviews the current understanding of intercontinental transport of gases and aerosols, including photochemical

oxidants, from Asia and resulting impacts on regional and global air quality and climate. Important uncertainties regarding long-range transport of pollutants are also discussed.

## 2. Emissions and transformations of gases and aerosols in Asia

A thorough understanding of air pollutant concentrations and fluxes being affected by long-range transport requires accurate evaluation of pollutant emissions, transformations, and concentrations in the source regions. Accurate spatial and temporal emission data are necessary for modeling studies aimed at estimating the chemical transformations and resulting pollutant concentrations in the local as well as downwind areas. Some pollutants, e.g., ozone, are not directly emitted into the air but rather are produced from precursor interactions; and depending on their atmospheric lifetimes, the emitted and the produced pollutants from the source regions may or may not be transported over long distances. Atmospheric models can provide useful insights into the physical and chemical processes involved in these issues.

To better assess the regional and global transport of air pollution over Asia, a variety of emission inventories were developed recently based on approaches involving observations and numerical models. The basic method for developing such emissions inventories combines estimated emission-related activities for a given area and emission factors (in units of mass of emissions per unit of activity). The emission factors and activities are based on published analyses for the specific activities. In addition, since different activities vary greatly in emission type and quantity, emission measurements and pollution observations along with modeling calculations are used to crosscheck the emission factors, budgets and distributions in the inventory, e.g., the inversion method (e.g., see Streets et al., 2003).

### 2.1. Anthropogenic emissions

#### 2.1.1. Estimates of current emissions over Asia

During the past decade, there has been a significant effort towards improving the emissions estimations for Asia. As part of such analyses, measurement experiments combining aircrafts, satellite remote sensing, and ground stations have provided data that further help determine these emissions. These experiments include the aforementioned PEM-West, ACE-Asia, TRACE-P and PEACE. The integration of field campaigns and accompanied simulations over Asia provides a good way to reduce uncertainties in emission budgets and distribution, while also enhancing the ability to project plausible future trends.

Global (e.g., Cofala et al., in press), regional (e.g., Klimont, 2001) and national (e.g., Wang et al., 2005) inventories provide important information for estimating the Asian emission distributions. Particularly, many inventories for specified pollutants have been developed for the entire Asian continent or for specific countries (e.g., Corbett et al., 1999; Cofala et al., 2004). However, an integrated inventory for Asia is necessary for

understanding the total Asian contribution to long-range transport impacts. Recently, Streets et al. (2003) developed such an inventory for air pollutant emissions over Asia for the year of 2000 for atmospheric modeling studies and observation analyses performed as part of the TRACE-P and ACE-Asia experiments. Emissions were estimated for all major anthropogenic sources, including biomass burning, in 64 regions of Asia. The total annual Asian emission budgets were estimated as follows: 34.3 Tg SO<sub>2</sub>, 26.8 Tg NO<sub>x</sub>, 9870 Tg CO<sub>2</sub>, 279 Tg carbon monoxide (CO), 107 Tg methane (CH<sub>4</sub>), 52.2 Tg non-methane volatile organic compounds (NMVOC), 2.54 Tg black carbon (BC), 10.4 Tg organic carbon (OC), and 27.5 Tg ammonia (NH<sub>3</sub>).

Table 1 summarizes the emissions of each species in each Asian country based on the analyses of Streets et al. (2003). China dominates emissions of most species. It contributes 59% SO<sub>2</sub>, 42% NO<sub>x</sub>, 39% CO<sub>2</sub>, 42% CO, 36% CH<sub>4</sub>, 33% NMVOC, 41% BC, 32% OC, and 49% NH<sub>3</sub> out of the total Asian emissions. India is the second largest emission source and contributes 16% SO<sub>2</sub>, 17% NO<sub>x</sub>, 19% CO<sub>2</sub>, 23% CO, 31% CH<sub>4</sub>, 21% NMVOC, 24% BC, 27% OC, and 27% NH<sub>3</sub> out of the total Asian emissions. Other countries have much smaller emissions, though Japan is an important source of CO<sub>2</sub>.

Fig. 1 (from Streets et al., 2003) shows the relative contributions of various emission sectors to total emissions of individual species. It is seen that SO<sub>2</sub> and NO<sub>x</sub> emissions are mostly from industry, power generation, and transportation. By comparison, emissions of the primary aerosol species, BC and OC, are largely from power generation, residential (coal and biofuel) burning and biomass burning. Emissions of CH<sub>4</sub> and NH<sub>3</sub> are dominated by agriculture. This indicates

the importance of various source types in developing the emissions database is highly species dependent (Streets et al., 2003).

### 2.1.2. Uncertainties in emission estimations

Uncertainty estimations of an emission inventory are extremely important due to the fact that most emission inventories are generated based on estimations of emission factors and total emissions-related activities, which are usually limited by lack of information or reliable statistics, especially for a fast developing region like Asia. For some sources of emissions in some countries, very little is known about actual activity levels and/or emission factors.

Streets et al. (2003) integrated the emission uncertainties from different sectors based on the assumption that each sector was independent. Table 2 and Fig. 2 show their results for each species over seven prototypical Asian regions. It is found that emissions are known least well in India, the rest of South Asia, and Southeast Asia, mainly due to the large uncertainties in biomass burning and liquid fuel consumption in vehicles. Emissions are best known in Japan followed by the other East Asian countries. The overall uncertainty in all Asian emissions is as follows, ranked in increasing order of uncertainty at the 95% confidence interval: ±16% (SO<sub>2</sub>), ±31% (CO<sub>2</sub>), ±37% (NO<sub>x</sub>), ±65% (CH<sub>4</sub>), ±72% (NH<sub>3</sub>), ±130% (NMVOC), ±185% (CO), ±360% (BC), and ±450% (OC). For example, we are 95% confident that Asian emissions of SO<sub>2</sub> are within ±16% of the stated value. Note that a “±400%” confidence interval should be interpreted as “within a factor of five”, such that the true emission budget

Table 1  
Summary of national emissions of some important gases and aerosols in Asia for the year 2000 (Streets et al., 2003)<sup>a</sup>

Country	SO <sub>2</sub>	NO <sub>x</sub>	CO <sub>2</sub>	CO	CH <sub>4</sub>	NMVOC	BC	OC	NH <sub>3</sub>
China	20385	11347	3817	115749	38356	17432	1049	3385	13570
Japan	801	2198	1203	6806	1143	1920	53	74	352
Korea, Rep. of	829	1322	411	2824	1433	1161	22	28	172
Korea, DPR	227	273	120	3556	1345	234	22	106	98
Mongolia	101	221	69	2861	472	452	19	173	155
Taiwan, China	376	521	200	2127	560	510	8	10	152
Brunei	6	20	10	15	50	43	0	0	2
Cambodia	40	89	36	1707	708	305	14	89	86
Indonesia	884	1317	587	23105	6443	6903	206	1138	1390
Laos	21	96	44	2547	387	486	18	129	58
Malaysia	273	494	144	5552	869	1424	26	151	146
Myanmar	65	226	145	8446	2691	1671	65	421	341
Philippines	713	326	152	4102	2563	1398	36	192	273
Singapore	163	185	56	138	85	81	3	2	4
Thailand	961	1086	351	10815	3567	3052	72	364	388
Vietnam	193	283	169	9248	2907	1390	88	432	686
Bangladesh	140	220	123	4827	3608	819	52	268	763
Bhutan	6	8	4	172	42	36	2	12	10
India	5536	4591	1886	63340	32851	10844	600	2837	7399
Nepal	38	55	39	2087	917	346	21	135	168
Pakistan	1416	539	221	7076	5415	1344	85	368	1214
Sri Lanka	58	57	28	1348	407	275	11	55	92
International shipping	1083	1292	51	117	1	27	68	51	0
Asia total	34316	26768	9868	278564	106821	52150	2541	10420	27519

<sup>a</sup> Data are in Gg (Tg for CO<sub>2</sub>).

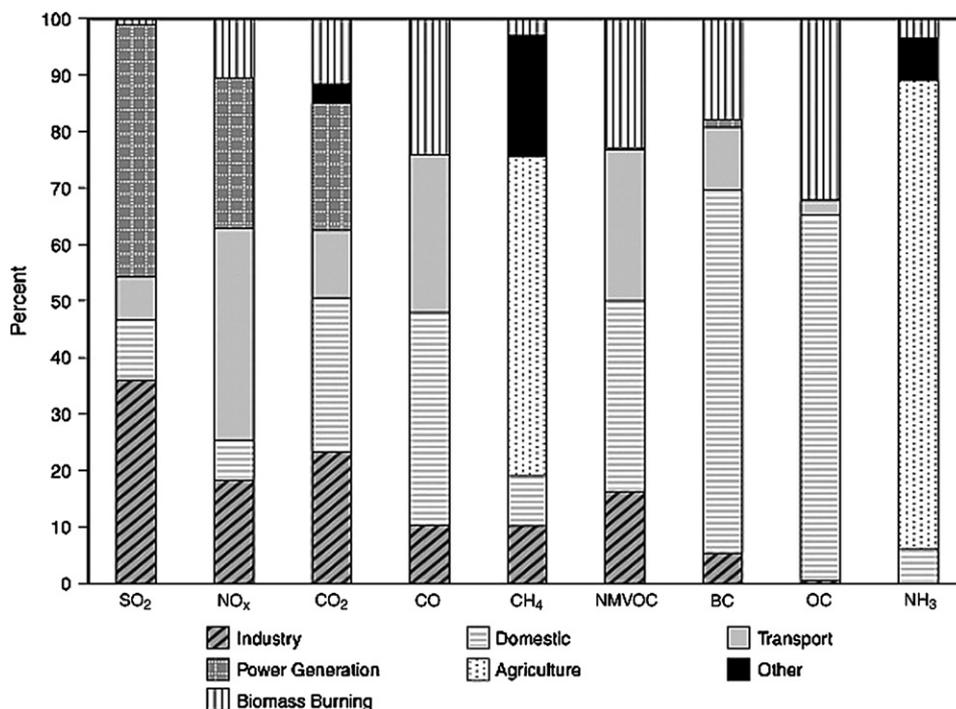


Fig. 1. Species emissions by emitting sector (Streets et al., 2003).

is within 20–500% of the mean at the 95% confidence level (Streets et al., 2003).

### 2.1.3. Future emission trend

There are two basic methods used to project future emissions: (1) Projections of activities that generate emissions (e.g., energy use, fertilizer use, livestock, and production of goods), together with the consideration of technology development (e.g., Streets et al., 2003); and (2) Projections of proxies, such as population or economic growth, and the resulting emissions over time, considering change of unit emissions with scenarios. Due to the rapid economic and social development in Asian countries, projections of activities are very difficult. Therefore, projections of proxies are more often used in estimating future trend of Asian emissions.

Asia is experiencing rapid population and economic growth. It is estimated that, by the year of 2010, over 4 billion people will be living in eastern Asia and the Indian sub-continent. Additionally, the Asian economy is growing at a phenomenal speed. For example, the Gross Domestic Product (GDP)

increased 9.5% per year in China and 7% per year in the Association of South East Asian Nations (ASEAN) countries from 1980 to 1990 (Hoffman, 1994). This rapid population and economic growth in many Asian countries has resulted in a significant increase in energy demand (Akimoto and Narita, 1994; Siddiqi et al., 1994). Presently, Asian energy demand is doubling every twelve years. Over 80% of all energy is derived from fossil fuels (Foell et al., 1995), with coal being the principal fuel and accounting for 76%, 54%, and 35% of the primary energy consumption in China, India, and South Korea, respectively (Shrestha and Bhattacharya, 1991). Further increases in energy demand are expected in future years if the industrialization of Asian countries continues without major advances in technology and energy efficiency.

This increasing energy demand concurrent with the increasing population/economic growth, if continued, would result in a large increase in pollutant emissions in Asia. Based on the current growing rate of emissions, Van Aardenne et al., 1999 estimated the Asian NO<sub>x</sub> emissions during the period of 1990–2020 under a no-further-control scenario and found

Table 2  
Uncertainty in pollutant emission estimates ( $\pm 95\%$  confidence intervals) (Streets et al., 2003)

Region	SO <sub>2</sub>	NO <sub>x</sub>	CO <sub>2</sub>	CO	CH <sub>4</sub>	NMVOC	BC	OC	NH <sub>3</sub>
China	13	23	16	156	71	59	484	495	53
Japan	9	19	7	34	52	35	83	181	29
Other East Asia	12	24	13	84	101	49	160	233	31
Southeast Asia	27	92	91	214	95	218	257	345	87
India	26	48	33	238	67	149	359	544	101
Other South Asia	35	63	44	291	109	148	379	531	101
International shipping	44	56	40	72	72	204	402	402	–
All Asia	16	37	31	185	65	130	364	450	72

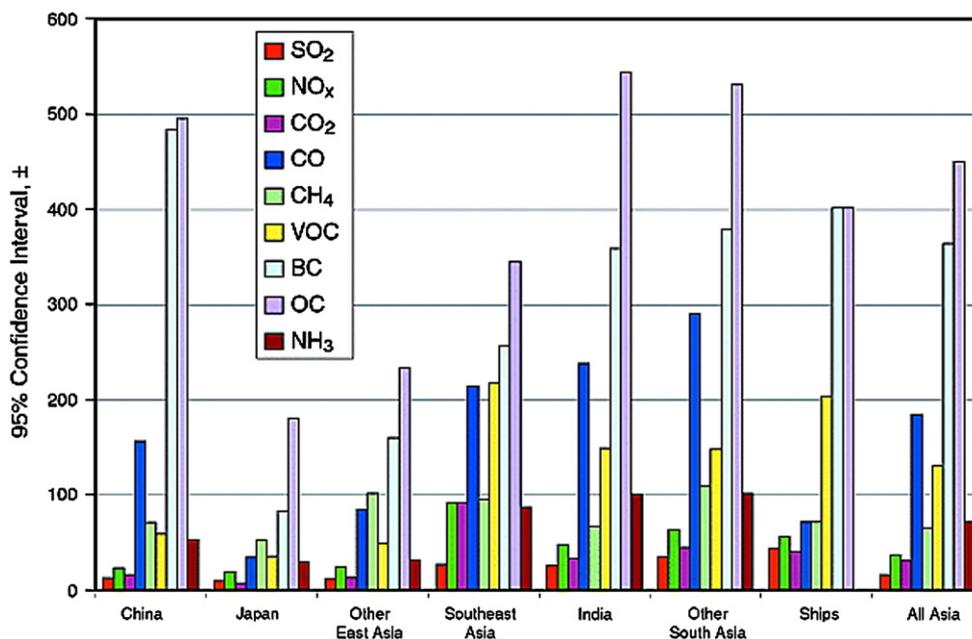


Fig. 2. Uncertainty (%) in pollutant emission estimates ( $\pm 95\%$  confidence intervals) (Streets et al., 2003).

that the  $\text{NO}_x$  emissions would increase by 350% to  $\sim 86 \text{ Tg NO}_2$  by 2020 (Fig. 3). Their results highlight the need to take the rapid growth in  $\text{NO}_x$  and other human-related emissions in Asia into account in studies of future air pollution and atmospheric chemistry not just in Asia but also in its downwind areas.

### 2.2. Natural emissions

Natural sources of atmospheric pollutants generally include dust emissions, living and dead organisms, lightning, and volcanoes. Over Asia, mineral dust is the major natural aerosol because of the vast desert regions. Other gases and aerosols

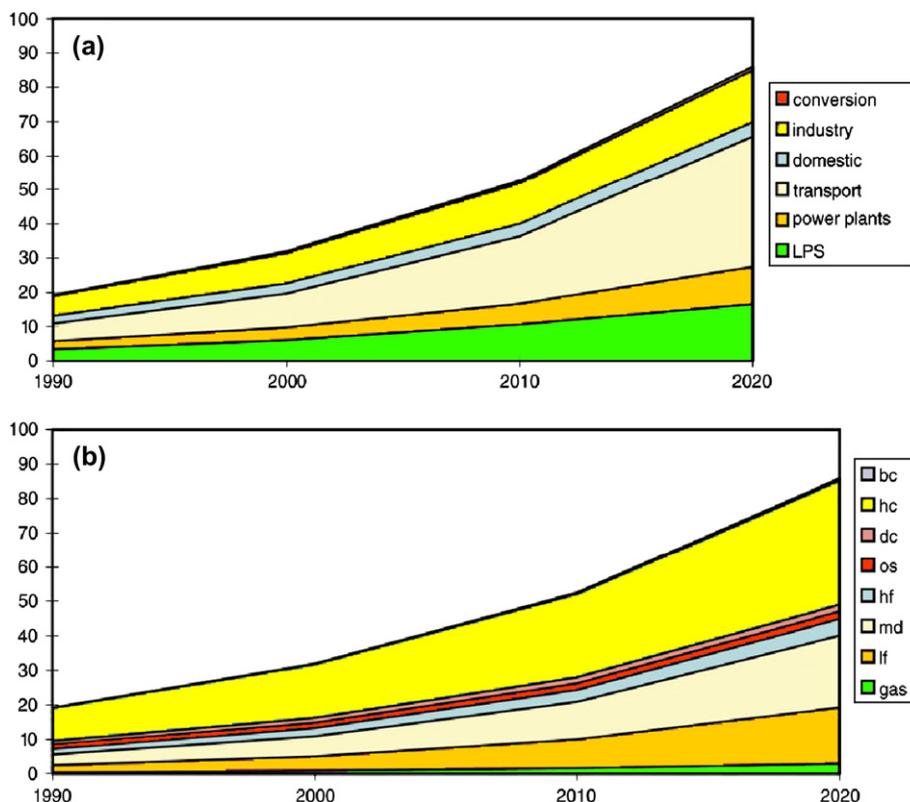


Fig. 3.  $\text{NO}_x$  emissions by economic and fuel type in the period 1990–2020 ( $\text{NO}_2 \text{ yr}^{-1}$ ) (Van Aardenne et al., 1999).

are mainly the result of anthropogenic emissions, or airborne in situ production, rather than natural emissions.

As the most important natural pollutant, dust aerosols play an important role in the climate system by affecting the radiation budget (e.g., Tegen and Lacis, 1996; Sokolik et al., 1998), biogeochemical cycles (Martin and Fitzwater, 1988; Martin, 1991; Archer and Johnson, 2000), and atmospheric chemistry (Dentener et al., 1996; Dickerson et al., 1997; Martin et al., 2003). Moreover, they have important consequences on surface air quality (Prospero, 1999). Dust aerosols originating from East Asia, one of the major dust emission regions in the world, may influence the ecological cycle of the North Pacific Ocean and the air quality over North America (Prospero, 1999). Many observation-based dust models have been developed to estimate the dust emissions from Asia. These dust emission models, whether based on parameterizations or physical mechanisms, are able to simulate dust emission processes to some extent after calibration using observation data. Tanaka and Chiba (2006) used a dust emission model to calculate the average emissions over Asia. They found that the emission budget and atmospheric burden of dust from East Asia were  $214 \text{ Tg yr}^{-1}$  and  $1.1 \text{ Tg}$ , which contributed to 11% and 6% of the global emission budget and burden, respectively.

Uncertainty assessments of natural emissions are more difficult than those of anthropogenic emissions, since the uncertainties associated with natural emission factors and emission algorithms are more difficult to quantify. For example, the dust emission process is very sensitive to changes in wind speed, land cover (e.g. vegetation type and density) and soil moisture (e.g. Mahowald and Dufresne, 2004; Guenther et al., 2006). Additionally, at the time scale of years to decades, it is also sensitive to ambient temperature and climate anomalies. Therefore an improved understanding of the processes controlling these factors is required for accurate estimate of natural emissions.

### 3. Observational evidence of intercontinental transport from Asia

#### 3.1. Long-range transport of ozone and precursors

The long-range transport of ozone and its precursors can have significant consequences on the dispersion of anthropogenic pollution and the global perturbation of tropospheric photochemistry, which can affect regional and national policies relative to air quality. For a given region, the concentration of tropospheric  $\text{O}_3$  is usually affected by two processes, namely: (1) in situ photochemical production from natural and transported anthropogenic precursors; and (2) export of ozone from other regions. The additional ozone either transported into the region or produced locally because of the long-range transport of precursors can add to concerns about the ability of that local area to reach specified air quality standards.

Recent measurements have provided much greater detail regarding the transport of  $\text{O}_3$  and its precursors from the source regions. Since  $\text{NO}_x$  is easily oxidized to  $\text{HNO}_3$

(the resulting lifetime of  $\text{NO}_x$  is approximately 1 day), it cannot be directly transported over long distances. Additionally,  $\text{HNO}_3$  in the troposphere is removed quickly by deposition and is not an effective reservoir for  $\text{NO}_x$ . However, research in the past decades has shown that peroxyacetyl nitrate (PAN) is a more efficient reservoir for  $\text{NO}_x$  in long-range transport (Hov, 1984; Staudt et al., 2003). So, while our particular focus in this section is on  $\text{O}_3$ , additional trace gases of interest include CO (lifetime 1–3 months), PAN (a product of  $\text{NO}_x$  photochemistry with a lifetime of weeks in the upper free troposphere), and various VOC species (lifetimes of hours to months).

#### 3.1.1. Evidence for transport of ozone and precursors from Asia

The ratio between concentrations of  $\text{O}_3$  and those of other trace gases can provide strong evidence for the impact of long-range transport on the ozone budget. CO is commonly used as a tracer for relevant human-related emissions in ozone transport studies. The  $\Delta\text{O}_3/\Delta\text{CO}$  ratio is widely used to examine the concentration variation of  $\text{O}_3$  along the plume of transported pollutants. Price et al. (2004) examine this ratio change along the transport plume from Asia to North America. Their analyses indicate that enhancement in  $\text{O}_3$  concentration accompanies the enhancement of pollution transport, thus verifying the transpacific transport of ozone and its precursors from Asia.

Several aircraft campaigns (i.e., TRACE-P, BIBLE, PEACE) over the NW Pacific, starting from the edge of the emission region, have characterized the Asian outflow of ozone and precursors (Kondo et al., 2002; Jacob et al., 2003; Parrish et al., 2004a,b). The PHOBEA aircraft campaigns over the northwest coast of the United States observed a number of transpacific Asian pollution plumes with elevated carbon monoxide, dust, and ozone (Jaffe et al., 1999, 2003a,b; Price et al., 2003; Bertschi et al., 2004). These experiments demonstrate the connections occurring between the emission region and the regions being affected by long-range transport processes, thus clearly showing that Asian pollutants contribute to the atmospheric composition over North America (Millet et al., 2004).

By tracking the formation of ozone, the pathway of the pollutants being transported is also determined. The ITCT 2K2 aircraft campaign, conducted in April–May 2002 off the California coast, provided a detailed chemical characterization of transpacific Asian pollution plumes including ozone, its precursors, and a number of other species, as well as the typical meteorological pathways for this transport (Parrish et al., 2004a). On 5 May, the NWS, 1996 WP-3D intercepted a “high-CO” Asian transport plume between  $33^\circ$  and  $37^\circ$  N around  $123^\circ$  W in an altitude band of 5–8 km (Fig. 4a and b). In this plume, CO levels peaked at 313 ppb, the highest CO concentration found in a plume seen during the ITCT 2K2 measurement campaign. In the lower part (5.5–6.5 km) of the enhanced CO altitude band, propane was also strongly enhanced. This suggested that there might have been multiple transport layers present from different anthropogenic sources

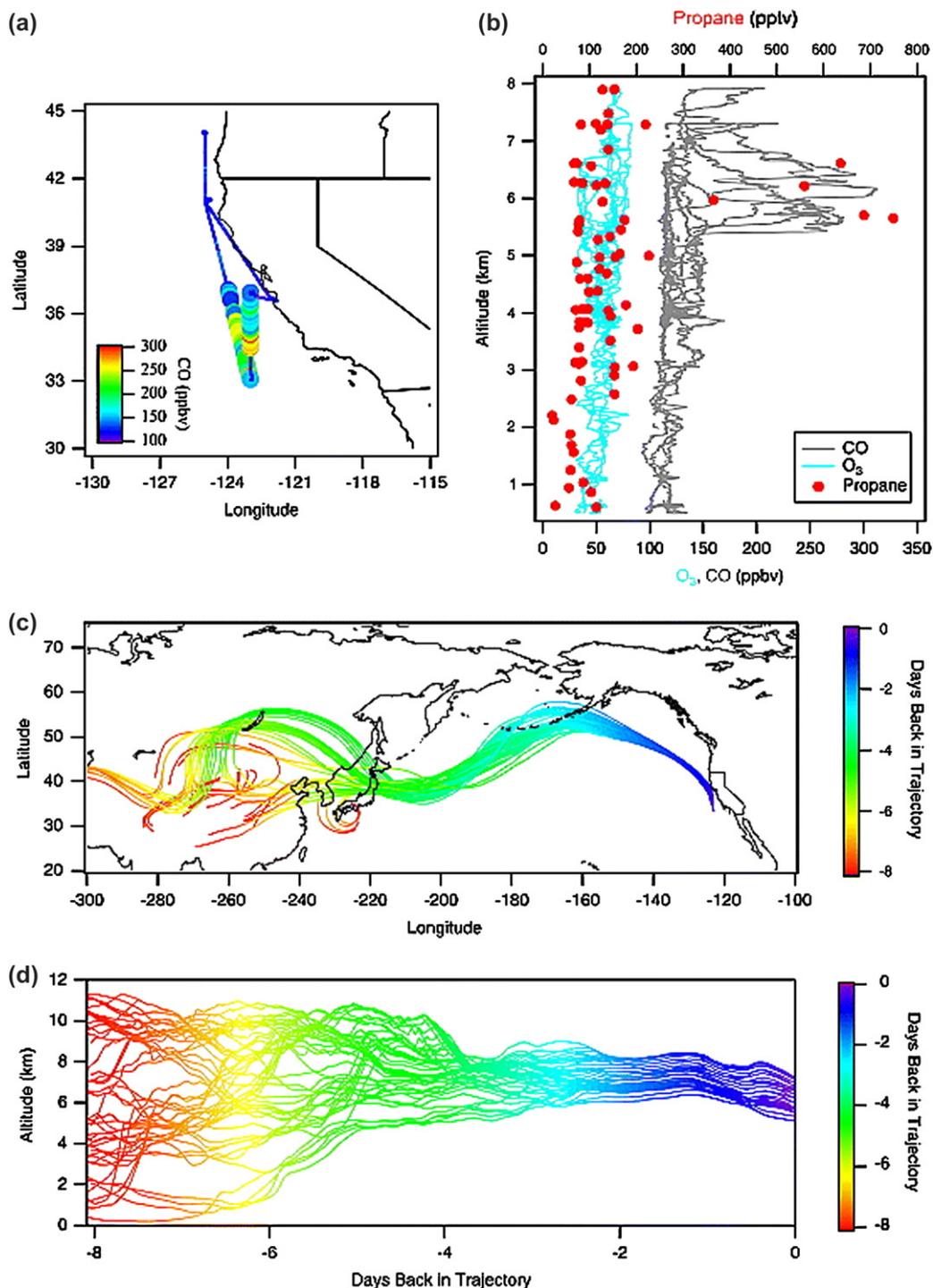


Fig. 4. (a) The 5 May flight track colored by the CO observations with the large markers, indicating the intersection of the transport plume. (b) Altitude (km) versus O<sub>3</sub> (teal) and CO (gray) on the bottom axis and propane (red circles) on the top axis above 0.5 km for the whole flight. (c) The 8-day back trajectories associated with the intersected plume colored by the number of days back along the trajectory. (d) Altitude of the back trajectories (Nowak et al., 2004).

(Brock et al., 2004). A detailed analysis of the meteorology associated with this transport plume is given by Cooper et al. (2004a,b). They found that the Asian pollution plume was transported through two warm conveyor belts (WCBs, i.e., the rising air streams transporting warm wet air from south-east to northern directions), which had developed over Asia

and the eastern Pacific. As seen in the back trajectories (Fig. 4c and d), the WCBs brought the plume from the eastern coast of Asia northward over the Aleutian Islands of Alaska and then southward toward the west coast of North America. Once lifted into the free troposphere, this plume was transported at altitudes greater than 4 km (Nowak et al., 2004).

### 3.1.2. Observations on photochemical oxidant transformations in the outflow

During the long range transport, ozone and its precursors experience complex chemical transformation in the outflow. Most soluble reactive odd-nitrogen ( $\text{NO}_y$ ) and other gas-phase species are affected by deposition processes as they are transported aloft from the continental boundary layer, especially in those regions where the environment is moist. PAN, however, is relatively insoluble and thus can be transported more efficiently with the lofted air plumes into the free troposphere where its lifetime is longer (weeks to months). When the pollution plumes eventually descend at far downwind distances, PAN decomposes to release  $\text{NO}_x$  and peroxyacetyl radical, which can lead to  $\text{O}_3$  production.

Observed  $\text{O}_3$  enhancements are derived from three possible sources: transport of  $\text{O}_3$  from the Asian boundary layer,  $\text{O}_3$  production in the outflow, and input from the stratosphere. Because it is difficult to evaluate the  $\text{O}_3$  production in the outflow, most analyses are based on the combination of model simulations and observations. Jaffe et al. (2003b) used aircraft and surface observations and the Naval Research Laboratory's (NRL) Aerosol Analysis and Prediction System to evaluate the impact of Siberian biomass burning emissions on the summer background CO and  $\text{O}_3$  of the west coast of North America, where the concentration of  $\text{O}_3$  was estimated from the model while the background  $\text{O}_3$  levels and transported  $\text{O}_3$  were determined from the measurements. The net result was that the long-range transport of precursors from Siberian forest fires increased surface ozone concentrations on the west coast of North America by about 15 ppb through chemical transformations.

### 3.1.3. Observations on impacts of transpacific transport

Ozone plays a key role in the troposphere due to its influences on human health, vegetation and climate. From the view point of the policymaker, a key issue is to determine what impact long-range transport has on local air quality. Since direct exposure to high concentrations of  $\text{O}_3$  can be harmful to human and ecosystem health and to crop production, most countries have defined limits on acceptable levels of ozone. The effects of long-range transport can be to add to the atmospheric burden beyond the levels of ozone produced locally, thus increasing the likelihood for surpassing the local ozone policy standards.

Many studies have focused on the impact of long-range transport on atmospheric composition over the west coast of the United States. Jaffe et al. (2003b) studied six episodes of trans-Pacific transport that occurred between 1993 and 2001 based on analyzing data from several ground sites in the Pacific Northwest and from aircraft observations in the region. Their results show that  $\text{O}_3$  was only enhanced in episodes that were transported in the free troposphere and in the absence of mineral dust, which means that transport in the boundary layer or transport of industrial emissions with mineral dust seems to preclude any significant  $\text{O}_3$  enhancement. A possible explanation is that heterogeneous reactions between dust and other chemicals reduce the concentrations of

various pollutants in the atmosphere reaching the receptor region. In summary, chemical transformations during the transport process can significantly influence the atmospheric composition.

In a longer time period analysis, Jaffe and Ray (2007) evaluated  $\text{O}_3$  data for the period 1987–2004 from 11 rural and remote sites in the north and western U.S., including two sites in Alaska. All sites show a seasonal cycle with a spring or spring-summer maximum. Seasonal and spatial patterns and long-term trends were identified. For most of the locations in the western U.S., they found significant inter-site correlations in the deseasonalized monthly means. This indicates that there are large scale factors that influence the monthly mean  $\text{O}_3$  concentrations across the western U.S.

## 3.2. Long-range transport of aerosols and their precursors

Atmospheric aerosols can either be emitted or be produced from gases emitted into the atmosphere. Primary emissions include industrial pollution, mineral dust, biomass burning and sea spray. The secondary aerosols are produced either from natural sources, e.g., for dimethyl sulfide (DMS) and biogenic hydrocarbons, or from human-related emission sources, e.g., for  $\text{SO}_2$ ,  $\text{NO}_x$ , and some hydrocarbons. Surface and aircraft in situ measurements, lidar measurements, and satellite remote sensing products have provided strong evidence for the long-range transport of aerosols from Asia.

### 3.2.1. Observations of pollutant aerosols in the outflow

Stationary observations can be used to examine the physical and chemical characteristics of transported pollutants. In Asia, dust and anthropogenic aerosols transported from China have been observed in Japan. Episodes of enhanced  $\text{SO}_2$  concentrations measured at the summit of Mt. Fuji, Japan (3776 m above sea level) provide an example of Asian outflow. Igarashi et al. (2006) found that the enhanced  $\text{SO}_2$  concentrations were always measured accompanied with enhanced CO and radon ( $^{222}\text{Rn}$ ) concentrations. Analyses on seasonal patterns in  $\text{SO}_2$  events showed a similar phase shift relative to the prevailing wind direction and relative humidity (RH) observed on-site. Their results indicate the importance of cloud processes in reducing  $\text{SO}_2$  concentrations within the pollution plume.

Aircraft measurements provide a useful way to examine the variation of atmospheric compositions in Asian outflow. Based on the Pacific Exploratory Mission Tropics (PEMT) A (1996) and B (1999) field campaigns, Moore et al. (2003) analyzed the aerosol chemistry and optical properties during the long-range transport of continental plumes over the Pacific Basin. In their study, a variety of anthropogenic and natural sources for these continental plumes were inferred from the data, including biomass burning, urban/industrial emissions, and dust storms. Additionally, aerosol size distributions (particularly for the refractory component) varied from one plume to another and most combustion-derived aerosols appeared

to be an internal mix of a refractory soot-like constituent in a volatile matrix.

### 3.2.2. Evidence for Asian aerosols and precursors transport over the Pacific Ocean

Measurement sites on Pacific islands can be strong indicators of long-range transport of aerosols and precursors. The transport of Asian anthropogenic aerosols across the Pacific was first documented in the 1980s from observations at island sites (Prospero et al., 1989; Prospero and Savoie, 2003; Ziemann et al., 1995; Arimoto et al., 1996; Huebert et al., 2003). These observations revealed a spring maximum in transpacific cyclone activity, coincident with the annual maximum in Asian dust emission, and the resulting dust signals in the observation sites. Aircraft observations in Asian outflow over the NW Pacific (Arimoto et al., 1997; Jordan et al., 2003; Maxwell-Meier et al., 2004) and the NE Pacific (Andreae et al., 1988; Clarke et al., 2001; Price et al., 2003) provided subsequent evidence of aerosol transport in the lower free troposphere, including coincident sulfate and dust.

More recently, satellite retrievals of aerosol optical depth (AOD) have been used to track Asian aerosol plumes across the Pacific (Husar et al., 1997, 2001; Edwards et al., 2004; Darnenova et al., 2005). For example, Darnenova et al. (2005) reconstructed the transport routes and coverage of dust episodes by using MODIS aerosol optical depth and TOMS aerosol index. Their analysis showed that, over the oceans, the presence of persistent clouds posed a problem in identifying the regions affected by dust transport, so only partial reconstruction of dust transport routes reaching the west coast of the U.S. was possible.

### 3.2.3. Observations on of the transpacific transport over North America

In situ aerosol measurements provide strong evidence that intercontinental transport of anthropogenic aerosol from Asia to North America and across the Atlantic Basin is a significant and consistent occurrence. Many studies on source-receptor relationships have clearly demonstrated that Asian pollutants account for at least some of the elevated aerosol events that are observed over the west coast of North America.

Evidence for aerosol transport from Asia to North America is summarized in Table 3. Table 3 is typically focused on episodic transport events that produce peak levels in measured surface concentrations over the west coast of North America. However, it shows that transpacific transport of aerosols does occur persistently from Asia to North America at small

background levels. The PM10 and PM2.5 in the table refers to particulate matter less than 10  $\mu\text{m}$  and 2.5  $\mu\text{m}$ , respectively.

Other evidence of intercontinental transport of aerosols from Asia to North America include pesticides in air masses from Asia in the Canadian Rocky Mountains, ecological indicators of possible trans-Pacific transport of POPs, and heavy metals in the Pacific Northwest and Arctic (Holloway et al., 2003; Wilkening et al., 2000). At Tagish in the Canadian Rocky Mountains, Bailey et al. (2000) detected elevated levels of pesticides during winter and spring and attributed them to pollution transported from continental Asia. A study on the Fraser River watershed in British Columbia concluded that toxic airborne pollutants from Asia might be a source of contamination in lake fish and sediments (MacDonald et al., 2000). Blais et al. (1998) found high POP concentrations in the snow pack of high mountains in the Canadian west. AMAP Assessment Report shows POPs and mercury in wildlife and human populations in the Arctic (AMAP, 1998).

## 4. Intercontinental transport pathways from Asia

The magnitude of long-range transport of pollution from Asia in affecting a given region is influenced by the meteorological conditions and their variability, the chemical and physical characteristics of the pollutants, and the strength and location of the emissions of the pollutants or their precursors. The Asian pollutant outflow is largely characterized by episodic events with clear seasonal variations. This section examines the major mechanisms and key characteristics of the transport pathways and resulting impacts on pollutant distributions.

### 4.1. Major types of transport pathways and mechanisms

The transport pathways from Asia are typically located in three distinct major regions, the tropics, the mid-latitudes, and the Arctic regions. In the tropics, the transport is mainly affected by rising motion from deep convection, poleward winds throughout much of the free troposphere with a return equatorward flow near the surface, and tropical easterly wind throughout the lower and mid-troposphere. At mid-latitudes, cyclones, along with the westerly zonal wind, and seasonal convection processes largely determine the transport pathways. In Arctic region, equatorward winds and the effects of the polar vortex largely determine the transport process.

A number of studies on intercontinental transport from Asia have shown various transport pathways in each of these three regions (Cooper et al., 2002; Stohl et al., 2002). The transport

Table 3  
Surface aerosol enhancements at U.S. from transpacific transport (Holloway et al., 2003)

Source region	Receptor region	Aerosol type	Aerosol enhancement ( $\mu\text{g m}^{-3}$ )	Reference
Asia (mean)	U.S. yearly means	Organic C Elemental C	0.013 (western U.S.), 0.007 (eastern U.S.) 0.005 (western U.S.), 0.003 (eastern U.S.)	Park et al., 2003
Asia (events)	Northwestern U.S., spring 1997	All	200 particles $\text{cm}^{-3}$	Jaffe et al., 1999
Asia (dust event)	Western U.S., April 1998	All	40–63 (PM10), 4–11 (PM2.5)	Husar et al., 2001; Vaughan et al., 2001
Asia (dust event)	Lower Fraser Valley, BC, Canada, 1998	All	18–26 (PM10)	McKendry et al., 2001

of photochemical oxidants and aerosols from Asia is of particular importance at mid-latitudes due to the strong emissions, high pollution levels and favorable atmospheric circulation patterns (e.g., cyclones). Generally, the following meteorological processes are critical in the long-range transport at mid-latitudes. (1) Persistent mid-latitude westerly result in intercontinental transport of Asian from west to east. (2) The high speed winds in the upper troposphere make it possible for pollutants to be transported rapidly. This is particularly important in summer, when the lifetimes of various pollutants, including ozone, are shorter due to the increased photochemical reactivity. (3) Lifting mechanisms are very important for pollutants to be transported into the mid and upper troposphere, where the intercontinental transport is more efficient than in the boundary layer. (4) Seasonal variation of wind speed and direction controlled by the changes in atmospheric circulation is responsible for the seasonal variability of pollutant transport pathways. Typically, stronger winds in the late-winter and early-spring along with modest photochemical reactivity result in the strongest intercontinental transport.

Liang et al. (2004) conducted a systematic analysis of the major meteorological mechanisms at mid-latitudes leading to Asian long-range transport, based on a combination of observations and model results. As a result, as shown by the schematic diagram in Fig. 5, outflow mechanisms over Asia are found to be affected by convective lifting (E1) and three transport pathways associated with mid-latitude cyclones: warm conveyor belt lifting (E2), postfrontal boundary layer transport (E3), and low-level prefrontal jet transport (E4). The possible import mechanisms over the Northeast Pacific are advection in the mean free tropospheric westerly flow (I1), boundary layer transport (I2), large-scale subsidence in the Pacific High (I3), subsidence in the dry air stream of a cold front (I4), and subsidence induced by mountain waves (I5) (Liang et al., 2004).

For most of the year, the mid-latitudes are dominated by mid-latitude cyclones with strong meridional stirring along tilted surfaces of constant potential temperature. Many studies have shown that air streams associated with mid-latitude cyclones are closely related to the pollutant pathways in Asia. Since the main long-range transport process is in the free troposphere, cross-boundary layer transport of pollution from the surface to the free troposphere (E2) is very important (Stohl, 2001; Cooper et al., 2002, 2004a; Hannan et al.,

2003; Liu et al., 2003; Miyazaki et al., 2003). The warm conveyor belt (WCB), the major air stream of mid-latitude cyclones, plays an important role in upward transport across the boundary layer (Carlson, 1998). The slow zonal flow in the boundary layer behind cold fronts (E3) is another major outflow pathway over Asia (Carmichael et al., 1998, 2003; Liu et al., 2003). In summer, convection systems (E1) are also very important for the uplift of Asian pollutants. And it is also an important outflow mechanism especially over Southeast Asia (Tropics Region) during the spring biomass-burning season (Bey et al., 2001). In addition, Liang et al. (2004) find that an additional outflow mechanism (particularly during seasons other than spring) is boundary layer transport in the prefrontal jet (E4), a band of rapid winds ahead of a surface cold front (Chen et al., 1994).

Mid-latitude cyclones tracking from west to east are the major mechanism for the export of aerosols and photochemical oxidants from the east coast of Asia throughout the year (Stohl et al., 2002). The warm conveyor belt, located on the eastern side of the cyclone and ahead of the surface cold front, is the most important air stream for rapid intercontinental pollutant transport because of its ability to lift boundary layer pollutants from the cyclone warm sector to the vicinity of the upper troposphere jet stream. Moving along the mid-latitude cyclone storm tracks, the jet stream can rapidly transport the pollutants downwind. Depending on the distance between pollutant sources and receptors, the journey of transpacific transport may take shorter or longer time, the latter in some instances requires two warm conveyor belts to finish the transport from Asia to North America (Cooper et al., 2004b).

To capture the pathway of pollutants in transpacific transport, Lagrangian trajectory models are widely used. Forster et al. (2004) forecast the pollutant pathway by using a CO trajectory model, which was based on the Aviation (AVN) Global Model (NOAA/NWS, 1996), for the Intercontinental Transport and Chemical Transformation 2002 (ITCT 2K2) aircraft measurement campaign. Fig. 6 shows the 54-h forecast of the total column of the Asian CO tracer from 15 May 2002, which follows a typical transport pathway for pollutants. It is shown that, because the transport from Asia to North America in this example is relatively slow, only patches of Asian pollution between 30° N and 40° N are predicted off of the North American West Coast.

In addition, there are related studies on the transport between the extra-tropics and the tropics (e.g., Pierrehumbert and Yang, 1993; Bowman and Carrie, 2002; Wei et al., 2002; Bowman and Erukhimova, 2004; Hess, 2005). Their results, however, show that the mass exchange between the extratropical and tropical regions is estimated to be one to two percent per day of the extratropical mass. In summary, the long-range transport in the tropics is generally not as important as that in the mid-latitudes (Bowman and Erukhimova, 2004).

In the tropics, Asian pollution can reach Europe via westward transport with the monsoon circulation from India to Africa and the Mediterranean (Lawrence et al., 2003). The overall impact of the summertime Asian Monsoon is to advect

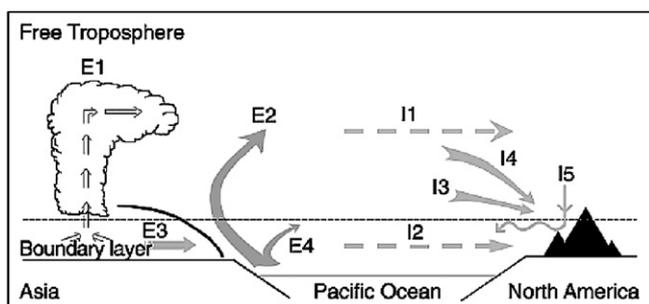


Fig. 5. Schematic of export mechanisms over east Asia and import mechanisms over the NE Pacific for long-range transport of Asian pollutants (Liang et al., 2004).

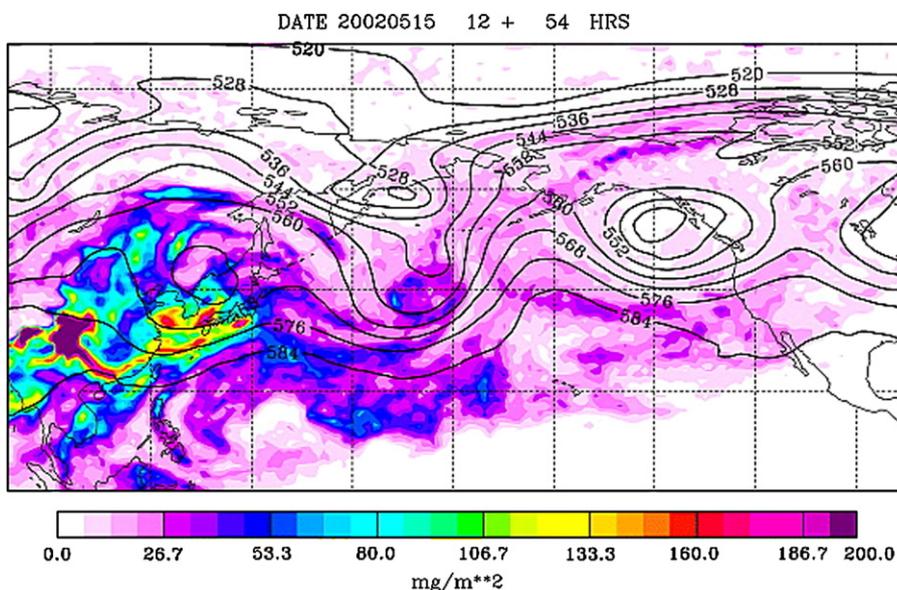


Fig. 6. Total column CO AVN-based forecast ( $\text{mg m}^{-2}$ ) of the Asian tracer for all age classes (0–20 days) from 15 May 2002 at 1200 UTC. Shown is the 54-h forecast (Forster et al., 2004).

relatively clean air from the Indian Ocean into southern Asia in the lower troposphere (Stohl et al., 2002), while the associated convection lofts anthropogenic emissions from northeast India and southwest China into the upper troposphere where they can recirculate above southern Asia within the semi-permanent summertime upper tropospheric anticyclone (Li et al., 2005). During winter the Asian Monsoon advects pollution from the Asian continent southward over the Indian Ocean (Lelieveld et al., 2001).

#### 4.2. Seasonality of intercontinental transport

Previous studies have indicated a clear seasonal variation in the intercontinental transport of Asian pollutants (Malm et al., 2000; Liu et al., 2003). This seasonality is particularly significant for aerosol transport due to the variations in Asian emissions and meteorological conditions leading to the long-range transport. The Asian aerosol emissions typically maximize during February–April since the strongest wind erosion in central Asian and biomass burning activities in Southeast Asia occur at this time (Duncan et al., 2003). In addition, meteorological fields have distinct seasonal characteristics, i.e., the westerly peaks in Spring. Liu et al. (2005) simulated the transport of nine continental tracers with uniform emissions and 2-week lifetimes using the global Model for Ozone And Related chemical Tracers Version 2 (MOZART-2) driven with the meteorological data from the National Centers for Environmental Prediction (NCEP) observation reanalysis during 1991–2001. Their results show that at mid-latitudes, the East Asian and Indian tracers have the largest transport potentials, particularly in spring.

The interannual variability of the transpacific transport potentials of most tracers is relatively high in winter and fall (particularly in February and September) but is low from April

to August (Liu and Mauzerall, 2005). Liu and Mauzerall (2005) showed that the average transport time from East Asia to the surface of western North America (including rapid episodic transport and slower pollutant outflow affecting the background tropospheric chemical compositions) for a pollutant with a atmospheric lifetime of 1–2 weeks (e.g., ozone) was 2–3 weeks in April, which was 1–2 weeks longer than the rapid plume transport. Holzer et al. (2005) quantified the transpacific transport from the industrialized regions of East Asia using a chemical-transport model, the results of which indicated seasonal-dependent transport pathways. Fig. 7 shows the peak times of the transit-time probability density function on the lowest model level together with the corresponding average air-mass fraction in a single region that has its last contact during the 24-h intervals 7, 14, and 21 days ago. As a result, the differences in climatological transport patterns in the four seasons are demonstrated in this figure.

Seasonal variability in pollution transport mechanisms around East Asia is strongly related to the monsoon system. For long-range transport, climate variations and meteorological condition fluctuations play important roles in determining the pathways and transformations of chemical pollutant plumes. In the early spring, the East Asian winter monsoon controlling the climate of the Asia continent reaches its maximum strength, characterized by strong wind speeds, persistent wind directions and low air temperatures below 700 hPa (Ding, 1994; Zhang et al., 1997). This, together with the northward moving warm air mass, leads to a strong instability in East Asia. As a result, mid-latitude cyclone activities become more frequent, a favorable condition for transpacific transport.

Another cause of the spring maximum in long-range transport from Asia, particularly for particles, is that the local emissions of various pollutants over Asia are the strongest in the Spring. Usually, the process driving the production of

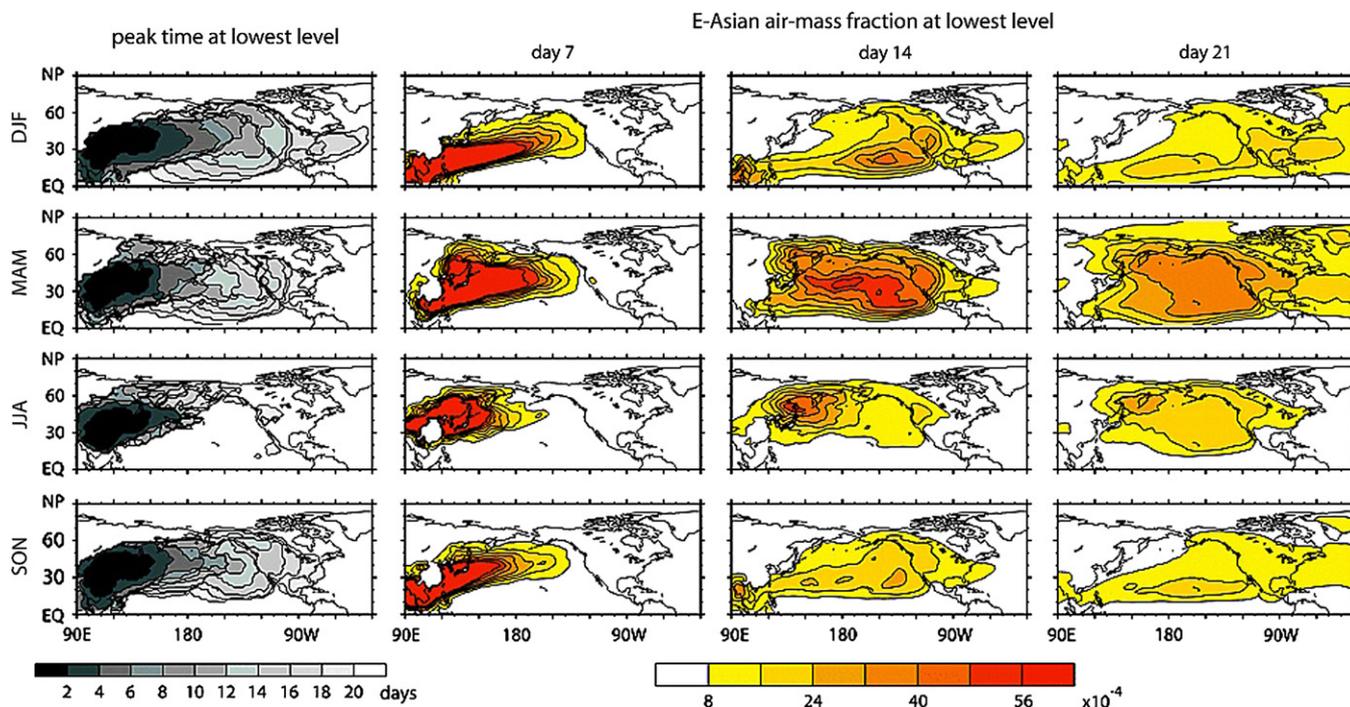


Fig. 7. Leftmost panels: The peak time of the climatological E-Asian air-mass fraction at the surface (lowest model level) for all four seasons, as indicated. Colored panels: The climatological air-mass fraction at the surface that had its last contact with the E-Asian source region during the 24-h periods 7, 14 and 21 days ago, as indicated for each season (Holzer et al., 2005).

atmospheric dust in Asia occurs mainly in the Spring, when the vegetation cover has not yet recovered. Anthropogenic emissions in springtime are also very strong due to increased biomass burning. The combined effects of the relative intensity of emissions and atmospheric circulation patterns thus appear to be responsible for the observed seasonal variability. A good example of the springtime Asian emissions effects is the occurrence of atmospheric brown clouds (Ramanathan et al., 2001a), where a haze layer is formed over much of southern Asia from December to April. It is found that this absorbing haze, which is mainly formed by black carbon and other aerosols, decreases the surface solar radiation and increases the lower troposphere solar heating; model simulations suggest the additional heating significantly perturbs tropical rainfall patterns (Ramanathan et al., 2001b; Ramanathan and Crutzen, 2003). One cause of this winter-spring time peak of brown clouds in southern Asia is that black carbon is mainly from coal combustion and biomass burning, with the former maximizing in winter in Asia due to power production for heating while the latter maximizing during February to April in Southeast Asia (Duncan et al., 2003).

The seasonality in the long-range transport from Asia leads to a seasonal characteristic of background ozone and aerosols over the west coast of North America. Analyses for the NASA Global Tropospheric Experiment (GTE) missions for the 1994 PEM-West B (Hoell et al., 1997) and the 2001 Transport and Chemical Evolution Experiment over the Pacific (TRACE-P) (Jacob et al., 2003) have shown an increasing amount of background ozone over the west coast of North America during spring (e.g., Jaffe et al., 2003a). The 2001 fourth Aerosol

Characterization Experiment (ACE-Asia) (Huebert et al., 2003) also has shown a spring time increase in aerosol levels over the same area. Studies of CO concentrations also indicate a similar seasonal trend (Liang et al., 2004). In addition, the Moderate Resolution Imaging Spectroradiometer (MODIS) satellite show the strongest transpacific transport occurring in spring at 40–55° N (Heald et al., 2006).

#### 4.3. Impact of pathways on pollutant distributions

The various transport pathways from Asia influence the distribution of pollutants reaching the receptor regions. Along individual air streams, pollutants may go to different regions before they are removed by wet and dry deposition processes. A major transport pathway for intercontinental transport from Asia is the westerly air streams along with warm conveyor belts exporting from Asia, where pollutants are transported in the lower and mid-troposphere above the eastern North Pacific (Heald et al., 2003; Nowak et al., 2004) and the U.S. west coast (Jaffe et al., 1999, 2003a; Jaeglé et al., 2003; Cooper et al., 2004a,b). On the other hand, boundary layer pollutants over source regions can also be lifted by convection into the upper troposphere before they are transported eastward; in this case, long-range transport occurs in the upper troposphere and seldom in the lower and middle troposphere. Modeling studies indicate that episodic long-range transport of CO from Asia to the northeastern North Pacific region occurs year-round roughly every 10, 15 and 30 days in the upper, middle and lower troposphere, respectively (Liang et al., 2004). This episodic transport is attributed to the frequency

of the cyclone activities at mid-latitudes. Also, the locations and properties of the South Asian brown clouds are determined by the transport pathways; because of the different pathways in the winter and summer time, the pollutants emitted over Asia go very different ways during the two seasons (Ramanathan et al., 2001b; Ramanathan and Crutzen, 2003).

## 5. Numerical modeling of intercontinental transport from Asia

Chemical-transport models (CTMs) have been used to explore intercontinental transport of pollutants from Asia in numerous areas, such as forecasting the transport events for field campaigns, post-campaign analyses of pathways/characteristics of pollutant transport and their effects on air quality and climate over downwind regions, and continuous simulations of the transport when field campaigns are not available. As the integrators of current knowledge, these models provide significant insight into the understanding of long-range transport processes and their importance. In this section, existing modeling studies related to the transpacific transport of aerosols and photochemical oxidants are discussed, focusing on the resulting impacts on atmospheric composition.

### 5.1. Transport of ozone and its precursors

#### 5.1.1. Surface ozone

Ozone is a major pollutant in the intercontinental transport research not only due to its adverse effects on air quality and climate over the downwind regions but also because of the complexity in the photochemistry involved in its production and destruction along the transport process. Recent surface ozone measurements in Asia in comparison with earlier measurements indicate that Asian ozone concentrations have increased significantly in the last few decades (Intergovernmental Panel on Climate Change IPCC, 2001), due to increases in anthropogenic emissions of ozone precursors in Asia. Many numerical models have been used to study the effects of Asian emissions on ozone concentrations in downwind areas.

The background surface ozone levels (i.e., average levels in marine air mass undisturbed by North American continental influences) in air plumes arriving at the west coast of North

America in spring have increased by about 30% over the past two decades (Jaffe et al., 2003a). Such a change in O<sub>3</sub> levels may indicate a profound change in the photochemical environment of a relatively remote portion of the troposphere. Although the ozone production off the coast of the Asia continent is rather weak in relatively fresh plumes (Tanimoto et al., 2005), substantial ozone production can occur in descending air masses before they reach the western U.S. and contribute to the violation of the National Ambient Air Quality Standards (NAAQS) under favorable meteorological conditions (Jaffe et al., 2003b). Heald et al. (2006) used satellite observations of carbon monoxide and aircraft measurements over the northwest Pacific along with a global CTM to quantify Asian pollution outflow and its transpacific transport during the spring of 2001. Their results suggested that PAN decomposition in trans-Pacific pollution plumes subsiding over the United States could lead to significant enhancements of surface NO<sub>x</sub> concentration and resulting effects to increase tropospheric ozone; in the transported plumes, the measured increasing trend in O<sub>3</sub> was highly correlated with the increasing trend in nitrogen oxide concentration.

A summary of published modeling studies on the estimation of surface O<sub>3</sub> enhancements in North America during different seasons resulting from intercontinental transport are presented in Table 4. The ozone enhancements are typically 1–5 ppb, but there exist occasions when the enhancements due to long-range transport can be as large as 10 ppb or more, and may exceed 30 ppb in future decades if the significant Asian emission enhancements continue (e.g., Yienger et al., 2000).

#### 5.1.2. Nitrogen species

Generally, for most locations, the amount of available NO<sub>x</sub>, which is largely emitted by combustion, microbial processes in soils, and lightning, limits the tropospheric production of ozone (Chameides and Stelson, 1992). As to the transpacific transport of Asian pollutants, a key issue is modeling the transformation of nitrogen species and the resulting production of ozone within the plumes being transported. A critical issue in relating NO<sub>x</sub> emissions to global ozone production is the fraction of emitted NO<sub>x</sub> that is ventilated out of the continental boundary layer (Jacob et al., 2003). The oxidation of NO<sub>x</sub> to nitric acid and PAN in the boundary layer takes place on

Table 4  
Estimated surface O<sub>3</sub> enhancements at U.S. in transpacific transport

Estimate region	Time	Source region	O <sub>3</sub> enhancement	Reference
U.S.	Monthly mean	Asia	1 ppb	Wild and Akimoto, 2001
Northwestern U.S.	Spring mean	Asia	4 ppb (mean); 7.5 ppb (max)	Berntsen et al., 1999
Northwestern U.S.	Spring mean	Asia	4 ppb	Jaeglé et al., 2003
Western U.S.	Spring mean	Asia	3–10 ppb (range during Asian pollution events)	Yienger et al., 2000
Western and Eastern U.S.	Monthly mean	Asia (1985–2010)	2–6 ppb (western U.S.); 1–3 ppb (eastern U.S.); highest April–June	Jacob et al., 1999
Western U.S.	Summer mean	Asia (1990–2020)	10–20 ppb	Yienger et al., 2000
U.S.	Summer mean	Asia + Europe	4–7 ppb (typical afternoon range); 14 ppb (max)	Fiore et al., 2002
Western U.S.	Spring mean	Global (1988–2002)	10 ppb	Jaffe et al., 2003a
Western U.S.	Monthly mean	Global (1987–2004)	5 ppb; 15 ppb at Summer	Jaffe and Ray, 2007

a timescale of a few hours. Nitric acid is removed rapidly by deposition. However, PAN (which is thermally unstable but not water-soluble) can be vented from the boundary layer into the mid and upper troposphere and transported globally under low temperature conditions, and eventually decompose to release  $\text{NO}_x$  as air masses subside (Jacob et al., 1999). Modeling studies show that the ozone production efficiency per unit  $\text{NO}_x$  consumed (OPE) increases as the  $\text{NO}_x$  concentration decreases (Liu et al., 1987), therefore the ozone production in the transported plumes is very efficient since the  $\text{NO}_x$  concentration is typically very low. Comparisons in the concentrations of nitrogen species in Asian and North American outflow indicate that 5–10% of the emitted  $\text{NO}_x$  over Asia is ventilated out of the boundary layer as PAN (Li et al., 2002; Koike et al., 2003; Parrish et al., 2004b). This PAN serves as the major means for Asian  $\text{NO}_x$  to be transported across the Pacific, with resulting ozone production in North America.

### 5.1.3. Carbon monoxide

CO, a product of incomplete combustion, can be effectively transported globally due to its long lifetime of 1–3 months in the troposphere (Staudt et al., 2001; Liu et al., 2003). A major source of the springtime Asian CO outflow is biomass burning in Southeast Asia, extending from northeast India to southern China and maximizing in Burma and Thailand (Heald et al., 2003). The air plumes from biomass burning are transported over the Pacific at lower latitudes than typical of other Asian anthropogenic pollutants (Heald et al., 2003).

Numerical models are used in studying emission and horizontal/vertical distributions in the transpacific transport of CO. Palmer et al. (2003a,b) utilized an optimal estimation inversion model, together with aircraft observations of continental outflow over the western Pacific in the campaign of the Transport and Chemical Evolution over the Pacific (TRACE-P) mission (March–April 2001), to improve the estimate of CO emissions from Asia. Their results showed significant Asian CO emissions from both biomass burning and anthropogenic sources. In addition, long-range transport simulations show that the transpacific transport of CO takes place at 25° to 55° N across the North Pacific Ocean in spring (Liu et al., 2003). Incorporating satellite observations and aircraft measurements over the northwest Pacific, Heald et al. (2006) used a global CTM to quantify Asian pollution outflow and its transpacific transport during spring 2001 and showed that anthropogenic CO was transported throughout the lower and mid troposphere.

## 5.2. Transport of aerosols and their precursors

Surface particulate matter (PM) concentrations over the U.S. can be enhanced by intercontinental transport of both natural and anthropogenic aerosols. However, different aerosol components from Asia have distinct physical and chemical characteristics, with different important consequences on their transformation and deposition during the transpacific transport processes. For example, depending on the size and chemical composition, the lifetimes of the aerosol particles in the

atmosphere vary from hours to several days; the lifetime affects the distance each kind of aerosol can be transported. Also, all aerosols are subject to dry deposition while only hydrophilic aerosols are subject to wet deposition. Therefore it is expected that during the transport process, the compositions of individual aerosols can be changed significantly. In the following sections, the modeled intercontinental transport processes for major types of aerosols are examined separately.

### 5.2.1. Mineral dust

As the major natural aerosol, the mineral dust is formed by the strong wind lifting of large quantities of dust particles and carried over long distances. A number of studies based on numerical dust emission and transport models have been conducted in various directions. For human life, recent modeling studies show that the fine dust, which can be transported across continents, has a particularly harmful effect on the human respiratory system (Dockery et al., 1993), and adverse impact on visibility (Malm et al., 2000). Scattering and absorption of solar radiation by dust have been verified to impact the Earth's radiation budget (Sokolik et al., 2001), the thermal structure of the troposphere, and actinic fluxes (Liao et al., 1999), and alter dynamical and photochemical processes through modeling studies. Additionally, the mineral dust in the troposphere can serve as condensation nuclei for cloud formation, which will further affect the radiation balance. Also, coating of dust particles under polluted conditions (Clarke et al., 2004) can change microphysical properties and promote surface chemical processes (Dentener et al., 1996; Jacob, 2000). Modeled dust minerals deposited to the oceans and to the terrestrial biosphere can act as a kind of fertilization and change the ecological cycle in the deposit areas (Prospero, 1996; Fung et al., 2000; IPCC, 2001; Meskhidze et al., 2005).

Recent modeling studies have verified that Asian dust aerosols can be transported across the Pacific Ocean and to the U.S. (Wilkening et al., 2000; Darmenova et al., 2005). Backward trajectory analyses and particle transport modeling suggest that dust aerosols originated from the Gobi and Taklamakan deserts of China and Mongolia contribute to air quality issues during Spring over the west coast of the U.S. (McKendry et al., 2001; Szykman et al., 2003). From model simulations together with the ground-based measurements from the Interagency Monitoring for Protected Visual Environment (IMPROVE) sites in the United States, Van Curen and Cahill (2002) found that Asian dust was pervasive over North America, extending beyond the sporadic springtime episodes.

The impact of Asian dust on the United States varies temporally and geographically. The Harvard group combined two dust mobilization schemes in the model and found persistent Asian fine dust in surface air over the western United States in Spring, with much weaker influence in Summer and Fall (Heald et al., 2006). They also found that the Asian influence over the eastern United States is 30–50% lower than found on the west coast (Fairlie et al., 2007).

The effects of atmospheric mineral dust is one of the most uncertain factors in determining the effects of aerosols on

global climate processes (IPCC, 2001). Recent studies of the contribution of dust to the climate radiative forcing show that the top of the atmosphere (TOA), net (shortwave plus longwave) dust radiative forcing can be positive (heating) or negative (cooling) depending on values of key variables (Liao and Seinfeld, 1998). Determining the sign and intensity of dust driven radiation forcing requires detailed knowledge of dust activity characteristics and spatial–temporal distributions (Tegen and Lacis, 1996; Alpert et al., 1998). Further study of the dust emissions generation mechanism in numerical models is needed for better estimation of the mineral dust distribution. Accurate spatial–temporal distribution of the transported dust is necessary to reduce the uncertainty in evaluating the impacts of dust on climate.

### 5.2.2. Anthropogenic aerosols

The social and industrial development of Asian countries over the past several decades has led to dramatically increased amounts of anthropogenic aerosols into the atmosphere (Kato and Akimoto, 1992; Van Aardenne et al., 1999; Streets and Waldhoff, 2000; Carmichael et al., 2002). Beyond the observation analyses, modeling studies provide more detailed information about the transport and physiochemical and optical properties of Asian aerosols. For example, Heald et al. (2006) used the global CTM, GEOS-CHEM, together with satellite (MODIS) observations of aerosol optical depths (AODs) over the North Pacific Ocean and surface aerosol measurements network over the remote U.S. regions (IMPROVE), to investigate the transpacific transport of Asian anthropogenic aerosol pollution and quantify its influence on U.S. surface aerosol concentrations. Distinct Asian sulfate episodes correlated with dust events are observed in the northwestern United States and simulated with the model. They found that the mean Asian pollution enhancement in that region in spring was  $0.16 \mu\text{g m}^{-3}$  with a 50% estimated uncertainty, which was higher than the estimated natural concentration of  $0.09 \mu\text{g m}^{-3}$  presently used as the visibility regulation objective in U.S. wilderness areas.

Sulfur dioxide is the most abundant precursor of anthropogenic aerosols. Anthropogenic emissions of sulfur oxides in Asian countries are of great concern because of their abundance as well as impacts on the atmospheric environment at regional and intercontinental scales. The regional budget and transport process of anthropogenic sulfur compounds over East Asia have been studied intensively using model simulations (e.g., Xiao et al., 1997; Bey et al., 2001; Tan et al., 2002). More recently, Tu et al. (2004) simulated the long-range transport of  $\text{SO}_2$  from east Asia to the central North Pacific troposphere, in comparison with the observations from transit flights. They found that the major  $\text{SO}_2$  removal processes appeared to be the heterogeneous process (i.e., loss to cloud droplets, rainout, washout) in the free troposphere and the loss to sea salt aerosols or ocean surface in the convective boundary layer. Therefore heterogeneous processes related to sulfur oxides are an important issue for troposphere chemistry and long-range transport research. This, however, has not been well studied.

## 6. Intercontinental transport impacts on the climate system

The transport of aerosols and photochemical oxidants in the atmosphere also has significant impacts on regional and global climate and marine ecological systems. The pollutants can scatter and absorb long- and short-wave radiations in the earth system and serve as nuclei for the formation of cloud, which affects the radiation balance of the atmosphere. When the pollutants deposit into the ocean, some of them can play a role as a kind of nutrient, which affects the marine biogeochemical cycle.

As a study of aerosol impacts on regional climate, Ramanathan et al. studied the formation of South Asian brown clouds, a result of long-range transport of Asian aerosols, and its impacts on the climate system. As was introduced in Section 4.2, brown clouds formed as a haze layer over much of southern Asia, which is mainly composed by black carbon. It plays a role to decrease the surface solar radiation and increases the lower tropospheric solar heating; model simulations suggest the additional heating significantly perturbs regional rainfall patterns (Ramanathan et al., 2001b; Ramanathan and Crutzen, 2003; Ramana and Ramanathan, 2006).

Recent work showed that the frequency of deep convective clouds along the North Pacific wintertime storm track increased from 1984 to 2005 (Zhang et al., 2007). Model simulations indicated that these increases in the deep convective clouds and the resulting increasing precipitation could be explained by aerosol effects associated with Asian outflow.

For the ocean system, some studies have shown that the transport of mineral dust is an important source for ocean iron. Since iron is an essential nutrient for all organisms, this dust deposition can cause fertilization effects on the ocean and thus impacts the oceanic primary production and  $\text{CO}_2$  uptake through the mechanism of “biological pump”. Jickells et al. (2005) have found that the temperate North Pacific was affected by the dust supply from the Asian deserts. This kind of effect will change the ocean currents in the Pacific. Furthermore, it will affect the regional and global climate system through the air-sea interaction processes.

## 7. Discussion and conclusions

The intercontinental transport of aerosols and photochemical oxidants from Asia is an important issue to downwind regions because of the concern that regional control strategies for air pollution may be inefficient if background concentrations of pollutants increase due to the rapid rising emissions and pollutant levels in Asia. Also, the Asian export of pollutants may have consequences on the regional and global climate system due to the influence on the atmospheric composition, radiation, cloud formation, and marine ecosystems.

The pathways and characteristics of transpacific transport of Asian pollutants and their impacts on the Pacific Ocean and North America are confirmed by recent field campaigns, satellite remote sensing, ground observations and chemical transport model simulations. This effect on other regions and

countries raises important issues relevant to the pollutant mitigation over the downwind regions and suggests the necessity of global cooperation on emission reduction aimed at successful domestic pollution control (e.g., Holloway et al., 2003).

Uncertainties regarding the pathways, characteristics and future changes of the intercontinental transport of Asian pollutants remain. First, observations of pollutant concentrations over Asian countries and reliable public statistics used to generate emission inventories are very limited. To improve the interpretation of observations and model simulations of the pollutant transport, more effort should be made to reduce the uncertainty in the emission inventory over Asia, acquiring more reliable statistics of the emission factors and activity levels (bottom-up method), and/or using alternate approaches such as inversion modeling (top-down method). Second, more attention should be paid to the characteristics of transport pathways and transformations and variations in the transported plumes to better understand the relationship between pollutant sources and receptors from air quality and climate perspectives, including the effects on the cloud processes and marine ecosystems. Third, current studies on the Asian pollutant export, especially the field campaigns, mainly focus on spring and summer. However, transpacific transport in other seasons are also important due to their effects on the background tropospheric constituents and climate/air quality issues in downwind regions. Also, the understanding of interannual characteristic of the long-range transport is still very poor. Multi-year continuous observations and simulations should be pursued in more detail in the future. Fourth, possible future changes in the long-range transport due to climate change and in response of changes in Asian emissions and resulting impacts on air quality over North America and regional/global climate require much more study. Future industrialization in Asian countries could lead to even more emissions and pollution over the source regions, although stricter environmental policies might lead eventually to pollution reductions over Asia. The efficiency of the pollutant transport could be changed under future climate conditions due to effects on transport pathways and on lifetimes of the gases and particles. A stronger research agenda to address these issues will likely be very important to policy-making processes aimed at regional pollution mitigation.

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