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Aircraft observations of dust and pollutants over northeast China: Insight into the meteorological mechanisms of transport

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[1] The meteorological mechanisms for lofting trace gases and aerosols out of the planetary boundary layer (PBL) into the free troposphere are key to understanding local air pollution problems as well as regional and global atmospheric chemistry and climate issues. Over the North American continent, convective storms and lifting in warm conveyor belts transport pollutants into the free troposphere. Little is known about the vertical distribution of pollutants and dust over east Asia, and the processes leading to transport, transformation, and removal of these species remain uncertain. To provide insight into these mechanisms, we report on eight flights based out of Shenyang in NE China as part of the U.S./China EAST-AIRE project conducted in April 2005. We evaluate profiles of trace species, along with back trajectories and satellite data, in the meteorological context of cyclonic systems. The warm-sector PBL air ahead of a cold front was highly polluted, while in the free troposphere concentrations of trace gases and aerosols were lower, but well above background; we measured ~300 ppb CO, ~2 ppb SO_2, ~70 ppb O_3, and ~ 8×10^{-5} m^{-1} aerosol scattering between ~1000 and 4000 m altitude. Satellite observations indicate that the entire plume contained almost 10^5 tons of SO_2 and that the gas decayed with a lifetime of 3–5 d. Roughly the same mass of aerosol was transported into the free troposphere. Over the east Asian continent, dry convection appears to dominate with warm conveyor belts first coming into play as the cyclonic systems move off the coast.

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1. Introduction

[2] Atmospheric chemistry and meteorology sometimes conspire to loft dust and pollutants out of the planetary boundary layer (PBL) into the free troposphere (FT), where they disperse over a large area. These processes have been studied extensively over the developed world, but in the coming decades, much of the world's economic growth and associated atmospheric emissions will come from the developing world, with east Asia as one of the major players [e.g., *Huebert et al.*, 2003; *Intergovernmental Panel on Climate Change*, 2001; *Q. X. Liu et al.*, 2005; *Ma and van Aardenne*, 2004; *Simpson et al.*, 2006; *Streets et al.*, 2003; *Streets and Waldhoff*, 2000; *Tanré et al.*, 2005; *Wang et al.*, 2005; *Zhang et al.*, 2004]. Trace gases and aerosols can have an impact beyond the local environment and human health. Several recent articles have documented the detection of emissions from Asia as far downwind as North America [*Allen et al.*, 2004; *Bey et al.*, 2001; *Carmichael et al.*, 2002, 2003; *Hannan et al.*, 2003; *Holzer et al.*, 2005; *Jaffe et al.*, 2003; *Liang et al.*, 2004, 2005; *Liu et al.*, 2003; *Liu and Mauzerall*, 2005; *J. Liu et al.*, 2005; *Mari et al.*, 2004; *Mauzerall et al.*, 2000; *Merrill et al.*, 1989; *Prospero et al.*, 2003; *Tanaka et al.*, 1980; *Uematsu et al.*, 2002; *Wang et al.*, 2006; *Yienger et al.*, 2000; *Zhang et al.*, 2004]. Similarly, emissions over North America can impact air quality over both the North Atlantic and Europe [e.g., *Dickerson et al.*, 1995; *Stohl et al.*, 2003] and Europe to Asia [e.g., *Akimoto*, 2003].

[3] The key issue in determining the large-scale impact of emissions is quick vertical transport up and out of the PBL. In the PBL near the Earth's surface, NO_2 and SO_2 are removed by dry deposition and the ozone and aerosols that they form are short-lived. In the FT, NO_x produces ozone more efficiently and more of the SO_2 is converted to the sulfate aerosol that impacts atmospheric radiative balance and cloud properties. Two mechanisms, both associated

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with wave cyclones, are involved in this quick transport [*Cooper et al.*, 2004; *Hess*, 2005; *Oshima et al.*, 2004]. Ahead of cold fronts, air can rise along lines of constant entropy in what has been called warm conveyor belt (WCB) circulation, and convection can cause rapid lofting of PBL air. Behind cold fronts, pollution levels tend to be low [see *C. Li et al.*, 2007], but mineral dust can be lofted in mesoscale wind systems [e.g., *Aoki et al.*, 2005], although convection may also be involved [*Jung et al.*, 2005].

[4] A global climatology of WCBs [*Eckhardt et al.*, 2004] indicated that lofting through this mechanism is common in winter at midlatitudes east of both North America and Asia, although Asian WCBs are less frequent and the local maximum lies farther off the continent. A model study of the relative importance of deep, moist convection versus nonconvective processes [*Hess*, 2005] showed that convection dominates in the summer. Neither study reported explicitly on the mechanism in spring, the aerosol transport season.

[5] Aircraft observations from TRACE-P [*Tu et al.*, 2003, 2004] showed substantial concentrations of SO_2 over the Pacific downwind of China. Cloud processing as well as exchange of air between the free troposphere and the marine boundary layer appeared to be involved, but chemical transport models had difficulty simulating the SO_2 distribution on cloudy days.

[6] Through a Chinese/American partnership among the Institute for Atmospheric Physics, Chinese Academy of Science, the Liaoning Provincial Meteorological Agency and the University of Maryland, we conducted intensive field campaigns on the ground and from aircraft during spring 2005 under the East Asian Study of Tropospheric Aerosols–An International Regional Experiment (EAST-AIRE) [*Z. Li et al.*, 2007]. We have developed a research platform for direct investigation of dust and pollutants in the source region. In this paper we present results from eight flights focusing on case studies where vertical profiles of trace gases and aerosols are measured over NE China. These data provide insight into the meteorological mechanisms for lifting dust and pollutants to high altitude and long-range transport.

2. Experimental Methods

[7] The sampling platform used for this study was a twin engine turboprop Y-12 research aircraft, similar to a Twin Otter (Figure 1). The aircraft was outfitted with a suite of trace gas and aerosol instruments described previously [*Taubman et al.*, 2004]. The inlets were engineered onto the forward part of the upper fuselage, ahead of the engines. Trace gases were sampled from an aft-facing inlet while a forward facing, isokinetic inlet fed the aerosol instruments. Because of inlet line losses, sampling of super-μm particles is inefficient, and measurements are representative of sub-μm particles only. Temperature, relative humidity (RH), and pressure were measured using a thermistor, capacitive thin film, and Rosemount Model 2008 pressure transducer, respectively.

[8] Ozone data were acquired with a commercial instrument using UV absorption at 254 nm (Thermo Environmental, TEI Model 49, Franklin, MA), with 4 s temporal response. For observations of CO, we used a modified [*Dickerson and Delany*, 1988; *Doddridge et al.*, 1998; *Novelli et al.*, 1998] commercial (TEI Model 48) nondispersive infrared (NDIR) gas filter correlation analyzer, calibrated with CO working standards referenced to a National Institute of Standards and Technology (NIST) Standard Reference Material (1677c 9970 ppbv CO in nitrogen, certified; NIST, Gaithersburg, Maryland). A modified [*Luke*, 1997] commercial pulsed-fluorescence detector (TEI Model 43C) was used for measurements of ambient SO_2. Particle light absorption was measured using a Particle/Soot Absorption Photometer (PSAP, Radiance Research, Seattle, WA). The intensity of 565 nm light was quantified after it passed through a filter on which ambient aerosol was deposited [*Anderson et al.*, 1999; *Bond et al.*, 1999]. This instrument developed a problem with the flow monitor, and data will be reanalyzed and presented elsewhere [*Li*, 2007]. Aerosol scattering was determined with an integrating nephelometer (TSI Model 3563) that measured the total particle scattering coefficient (b_{sp}) at 450, 550, and 700 nm [*Anderson et al.*, 1996].

Figure 1. Y12 Chinese twin engine turboprop research aircraft used for the studies described here. Operated by the Liaoning Provincial Meteorological Agency, this aircraft carried the instrument package shown in the insert. Inlets, not shown, were mounted on an overhead aperture in the fuselage ahead of the wings.

[9] The aircraft campaign was conducted in April 2005 out of Shenyang, the capitol of the province of Liaoning in NE China, about 650 km NE of Beijing. Shortly after the ground-based campaign in Xianghe [*C. Li et al.*, 2007], we flew a total of eight research missions on 1, 5, 6, 7, 9, 10, 11, and 12 April. The flights were confined within the province, a radius of about 500 km, but covered the majority of major emission sources such as the heavy-industry cities of Shenyang, Fushun, Tieling. The flight routes and additional observations can be viewed from the EAST-AIRE homepage:http://www.atmos.umd.edu/~zli/EAST-AIRE/station.htm.

[10] Flights described here include several profiles over the airport and to the north and south near the cities of Liaozhong and Tieling. At Liaozhong, a ground-based observation station was installed to measure aerosol, radiation, and cloud quantities in support of the air campaign [*Xia et al.*, 2007]. This paper presents summary data from all eight flights with a focus on results from ahead of a cold front (5 April 2007) and behind it on (7 April 2007).

[11] The OMI instrument measures the column content of atmospheric SO_2 using UV spectroscopy [*Krotkov et al.*,

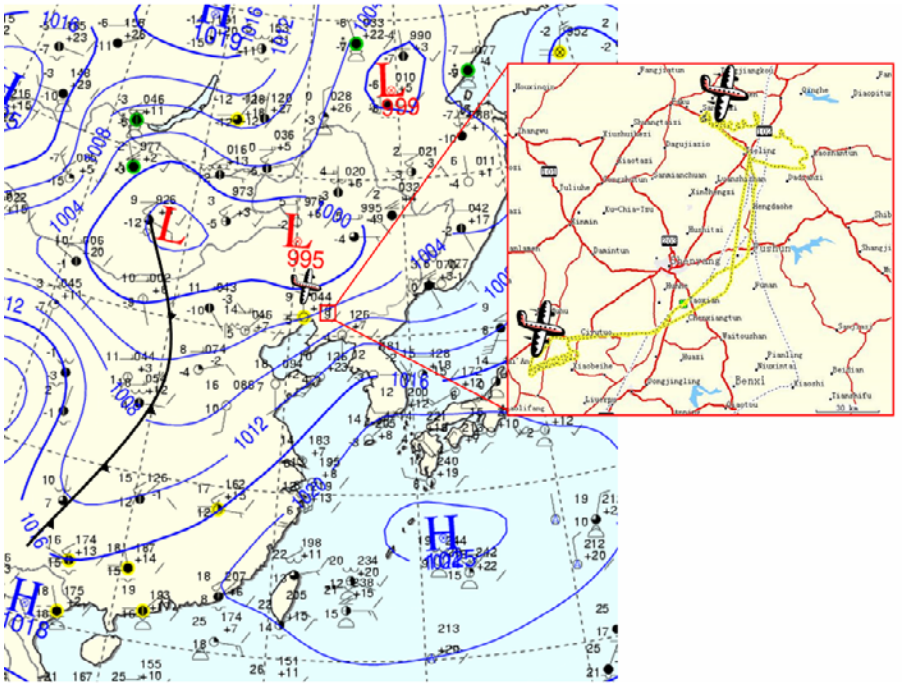

Figure 2. Surface analysis (Korean Meteorological Association) for 0000 UTC 5 April 2005 showing a low-pressure system (cyclone) moving toward Shenyang. The approximate location of the flights is shown by the image of an aircraft, and the actual flight pattern is shown in the insert. Altitude profiles were conducted north and south of Shenyang (locations marked with aircraft symbols). Note little cloud cover or precipitation was reported in the vicinity of the flights.

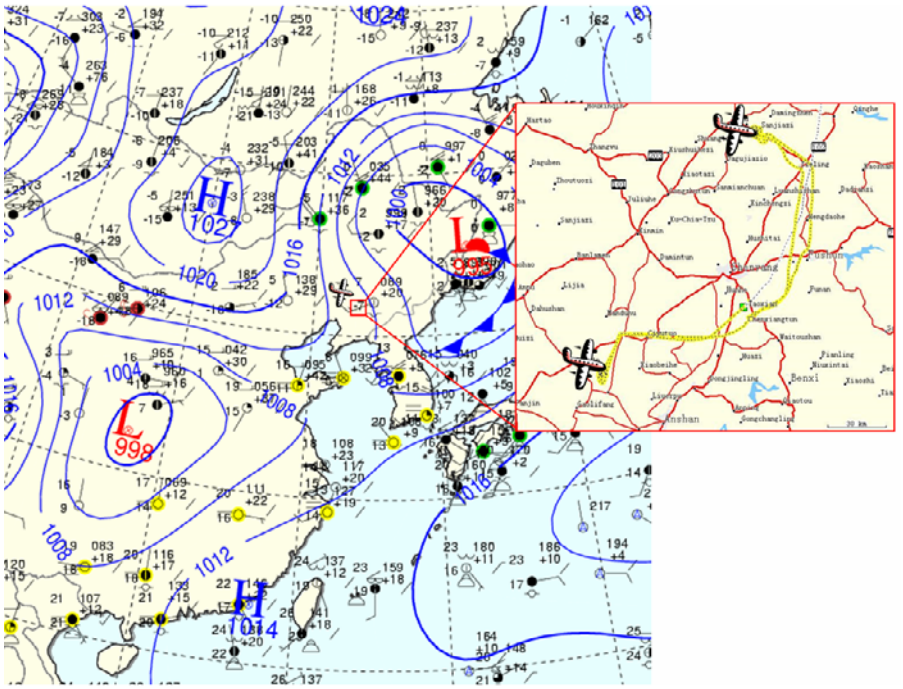

Figure 3. Surface analysis (Korean Meteorological Association) for 0000 UTC 7 April 2005 showing a cyclone NE of Shenyang. The flight was conducted behind the cold front in northerly flow. Clouds and precipitation (green background on station symbols) were reported north of the flight. The approximate location of the flights is shown by the image of an aircraft, and the actual flight pattern is shown in the insert. Altitude profiles were conducted north and south of Shenyang (locations marked with aircraft symbols).

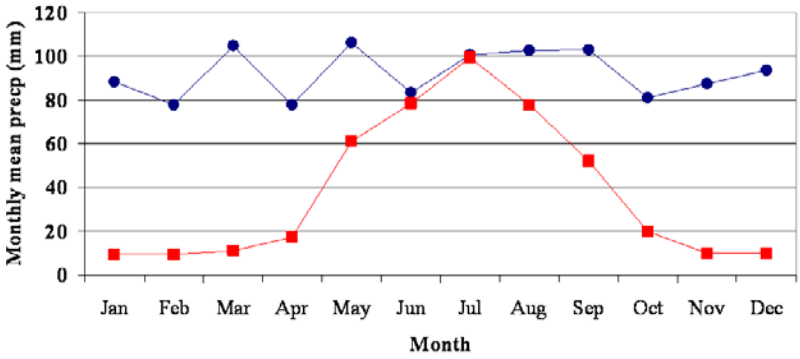

Figure 4. Climatology of precipitation near Baltimore and Beijing [*Arakawa*, 1969; *Bryson and Hare*, 1969]. Note that most of the rainfall over NE China comes in the summer while over the NE United States it is more evenly distributed. In the spring, frontal passages and convection are common over both continents, but little rain is produced over NE Asia; thus dry convection may play a larger role in pollutant transport. NE China has experienced severe drought recently making the differences even greater [*Zou et al.*, 2005].

2006]. Because much of solar UV radiation is scattered by the atmosphere before reaching the ground, OMI is more sensitive to SO_2 at high altitudes, and the concentration retrieved depends on the vertical profile assumed. The OMI default SO_2 retrieval algorithm assumes that the mixing ratio falls off with a scale height of about 3 km; the retrieval is improved with the observed altitude profile as described below. The uncertainty in the SO_2 column content retrieved from OMI depends on several factors including instrument noise, signal averaging, and the altitude of the target gas. As described in detail by *Krotkov et al.* [2007] when averaged over an area of 5° longitude × 5° latitude the total uncertainty is thus about 35% with 95% confidence for the mass of SO_2 observed on 5–7 April 2005; precision is better.

3. Meteorology and Climatology

[12] The flights took place between 1 and 12 April 2005, and during this time two major cyclonic systems traversed the experimental region in NE China, with cold fronts approaching from the NW on 5 April (Figure 2) and from the SW on 9 April. Behind these fronts the area around Shenyang was generally under the influence of high-pressure systems (Figure 3; see http://abc-gosan.snu.ac.kr/ABC-EAREX2005).

[13] Lofting over North America and resulting interhemispheric transport has been studied more thoroughly than over China and a comparison of the two continents can be enlightening. Cold front passage is prevalent over both continents, but spring in northern China is dry compared to North America (Figure 4). Over North America, cold fronts often demark the collision of maritime tropical (mT) and continental polar (cP) air masses, resulting in convective clouds and precipitation. Over northern China, air in the warm sector has generally not been over water long enough to develop maritime character; the low humidities inhibit precipitation. Most of the precipitation in NE China falls in the summer months (June to August) while precipitation in the NE United States falls more uniformly throughout the year. The area around Shenyang receives less than 20 mm of rain per month in March and April, while Baltimore, for example, receives about 80 mm per month. The total annual precipitation over NE China is about half that over the NE United States (Figure 4).

[14] In contrast to precipitation, the total average cloud cover over NE China is not dramatically different from that over the NE United States. Over midlatitudes (35 and 45°N) the average total cloud cover in spring (MAM) is about 50% near and upwind of Beijing (115 to 130°E) and 64% over the American Northeast (70 to 85°W). Cumulus clouds were reported about 7% of the time over the NE China and 9% over the NE United States. The peak in the diurnal cycle at both locations falls at 1300 to 1400 LST; cumulonimbus clouds are present in about 1% of the observations of both continents. Aerosols seem to have reduced cloud

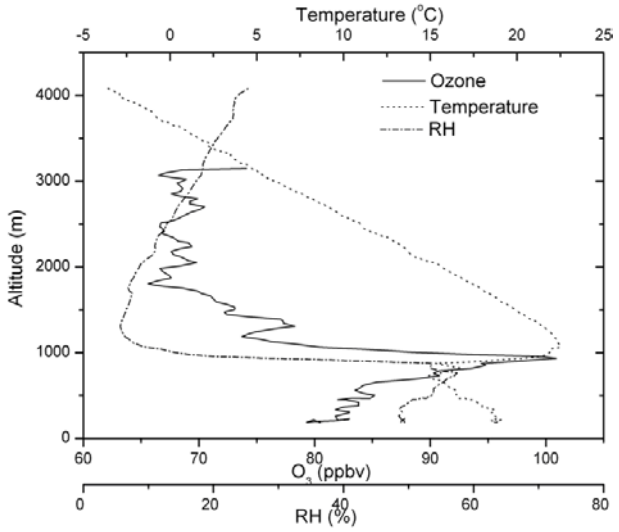

Figure 5. Altitude profile of ozone, RH, and temperature measured south of Shenyang at 1100 local time on 5 April 2005. Note maximum in RH and ozone near the strong temperature inversion (7°C) near 1000 m altitude. In the lower free troposphere the air is dry and ozone concentrations are lower. The other three profiles on this flight show similar results; see http://www.atmos.umd.edu/~zli/EAST-AIRE/station.htm.

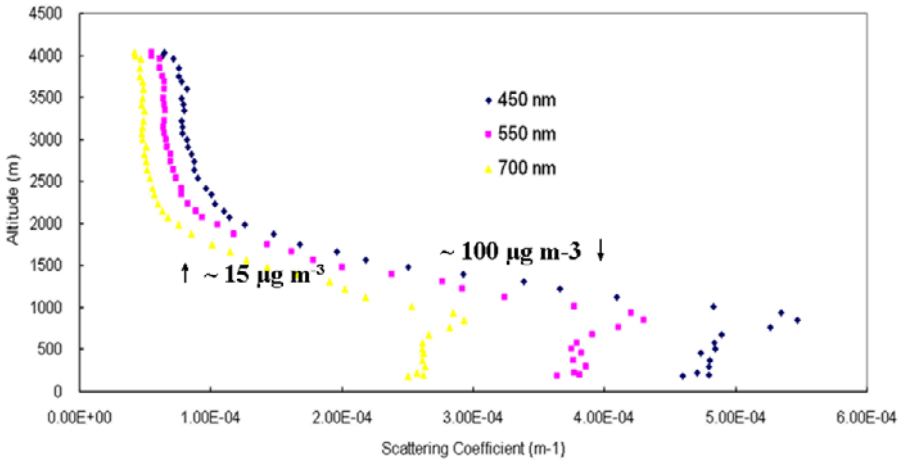

Figure 6. Altitude profile of aerosol scattering coefficient, b_{sp}, for the morning (~1100 LST) flight of 5 April 2005. Strong scattering in PBL reflects the heavy aerosol loading (~100 μg m^{-3} for a scattering efficiency of 4 m^2/g). Note rapid drop in scattering above the inversion (Figure 5) but the absolute value of b_{sp} remains high. Integrating the b_{sp} over altitude yields an aerosol optical depth near unity at 550 nm. MODIS (see EAST-AIRE website) detects AODs of this order.

cover over China over the past few decades, but not enough to change the basic climatology [*Qian et al.*, 2006; *U.S. Department of Energy*, 1986]. In short, convective clouds form over China, but in spring little rain falls.

4. Results

4.1. Ahead of the Cold Front: 5 April 2005

[15] Substantial pollution was found aloft on 5 April 2005 when the aircraft flew ahead of a cold front (Figure 2). The aircraft flew out of Shenyang, capital of the Liaoning province, home to over six million inhabitants, and the heart of China's industrial northeast. Sizable concentrations of trace gases and aerosols were observed not only in the PBL, but also in the FT (Figures 5–8). Here we investigate the pollutants found at high altitudes, the pollutants that can have large scale impacts.

[16] Ozone mixing ratios (see Figure 5 for a representative example) reflect moderate photochemical smog formation, and show a peak just above the inversion. The scattering coefficient, b_{sp}, also shows a maximum at that

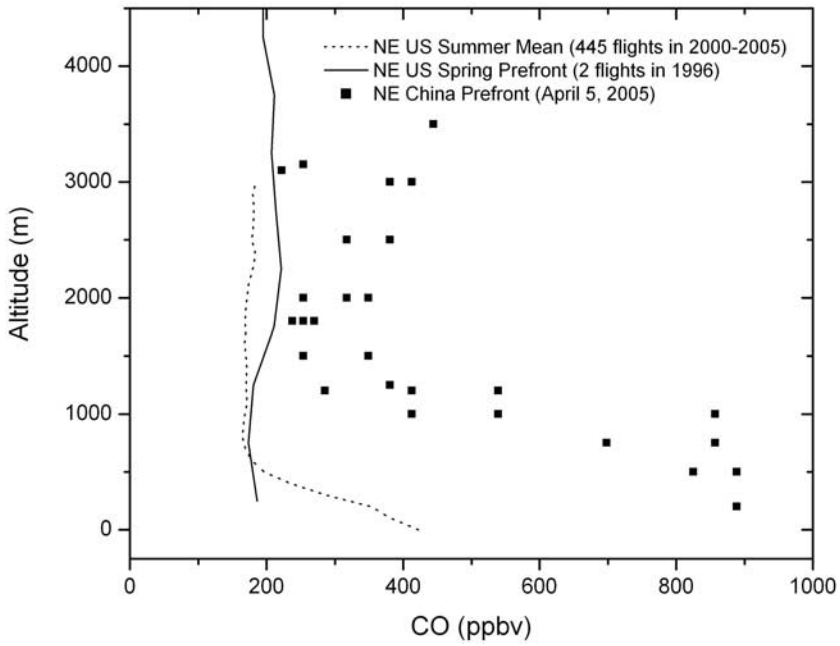

Figure 7. Altitude profile of carbon monoxide for all four profiles flown on flight of 5 April 2005 (squares). Mixing ratios drop off sharply above the inversion near 1000 m, but remain well above the background. The mean CO profile from ~400 flights made on smoggy summer days between 2000 and 2005 over the NE United States [*Taubman et al.*, 2006] is shown for reference. The standard deviation of CO for the data over the United States is less than 50 ppb.

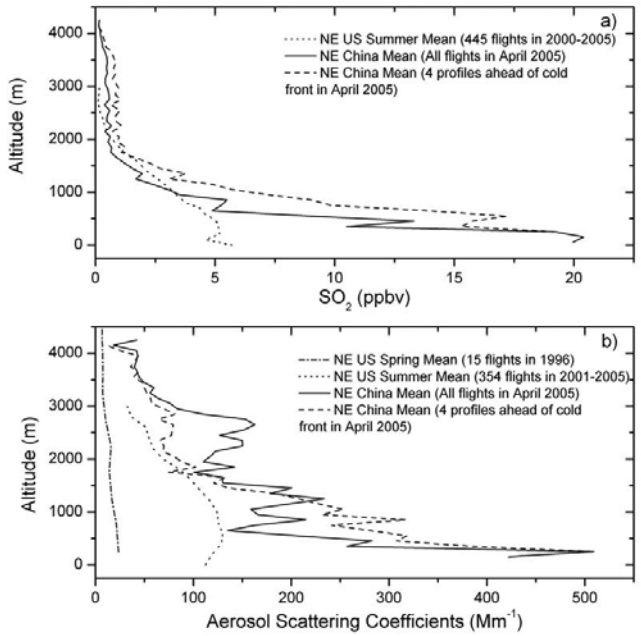

Figure 8. (a) Mean altitude profiles of sulfur dioxide for flights ahead of the front (dashed line) and for all eight flights (solid) conducted over China. Concentrations of SO_2 are highest at all altitude for profiles flown ahead of the front where mixing ratios drop off sharply above the inversion near 1000 m, but remain well above the background. For comparison the mean SO_2 profile measured on smoggy summer days over the NE United States [*Taubman et al.*, 2006] is shown as a dotted line. Concentrations of SO_2 over China are greater than over the United States at all altitudes; above the PBL where long-range transport is efficient, concentrations over China were about four times those seen over the United States. (b) Mean altitude profiles of optical scattering (b_{sp}) at 550 nm ahead of the front (dashed line) and for all eight flights (solid line) conducted over China. Scattering is stronger in the PBL for profiles flown ahead of the front because of high levels of pollution. Scattering is stronger aloft in profiles flown behind the front most likely because of wind-blown dust. For comparison, the mean b_{sp} profile measured over the NE United States in spring 1996 is shown as a dot-dash line and the mean for smoggy days is shown as a dotted line [*Taubman et al.*, 2006, also manuscript in preparation, 2007]. Concentrations of aerosols over China were greater than over the United States at all altitudes.

altitude (Figure 6); for reference a b_{sp} of 4×10^{-4} m^{-1} corresponds to 100 μgm^{-3} fine particulate matter for a scattering efficiency of 4 m^2 g^{-1}. Multiphase reactions in the aerosol rich PBL may inhibit ozone formation [*Lary et al.*, 1997], while a layer of aerosols that scatter UV radiation can inhibit photochemical ozone formation at low altitudes and accelerate it toward the top of the aerosol layer [*Dickerson et al.*, 1997]. These processes may contribute to the increase in ozone from the surface to 1000 m altitude. Concentrations of aerosols and trace gases fell off sharply above the strong inversion (7°C at 1100 LST), but remained well above background values. These heavy loadings of pollutants and possibly mineral dust persisted to the highest altitudes sampled, ~4000 m. The aerosols detected on this flight were widespread and impacted a large area of China, Japan, and the western Pacific on subsequent days (see NASA AI images http://jwocky.gsfc.nasa.gov/aerosols/aerosols_v8.html, MODIS image http://modis-land.gsfc.nasa.gov/ and the EAST-AIRE web site). These satellite observations show that this case study was an example of long-range transport of substantial concentrations of particulate matter.

[17] The meteorological conditions for this flight (Figure 2) involved low-level flow from the southwest, over some of the most highly populated and polluted parts of China. Winds veered with altitude becoming westerly in the midtroposphere. The low-level air from the warm sector ahead of the cold front brought heavy loadings of pollutants and dust (Figures 5–8) to the region around Shenyang. In the area of the flights and to the south few clouds were observed; the system brought little moisture or rain to the flight region until 6 April (flight discussed separately by [*Li*, 2007]), and by 7 April only cirrus clouds were observed.

[18] To help identify the meteorological mechanism responsible for the high concentrations of aerosols and trace gases, consider the back trajectories [*Draxler and Hess*, 1998] seen in Figure 9. Flow at low levels was from the southwest while flow at higher altitudes originated from the west. Little evidence of upward motion is seen in the prefrontal zone along the east coast of China; in fact HYSPLIT (Hybrid Single-Particle Lagrangian Integrated Trajectory) indicates a weak upward motion, suggesting that the WCB was not effective in lofting pollutants over the continent in this case study. The back trajectories employed here cannot resolve small-scale vertical motions such as those caused by convective clouds. We examined GOES9 satellite infrared images for 3–5 April 2005 for clouds, and

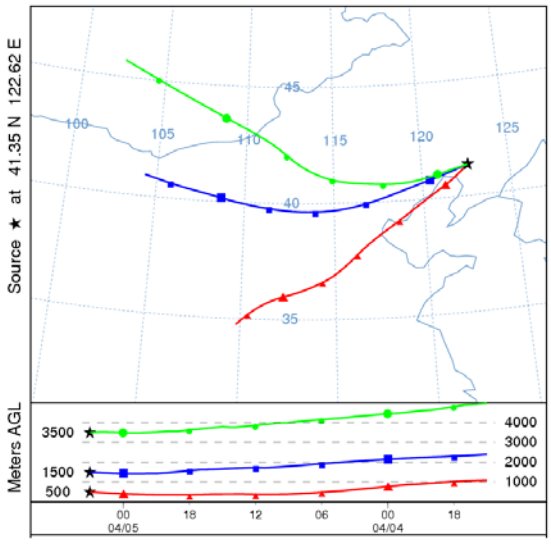

Figure 9. Back trajectories (36 h) calculated with HYSPLIT for air arriving at location of southern spiral (Figure 2) in the PBL (500 m above ground level) and in the FT (1500 and 3500 m). The plot at the bottom depicts vertical motions and shows little evidence of WCB lofting in the prefrontal zone for this episode. Back trajectories with end points at the second spiral, 300 km north and 2 h later are similar.

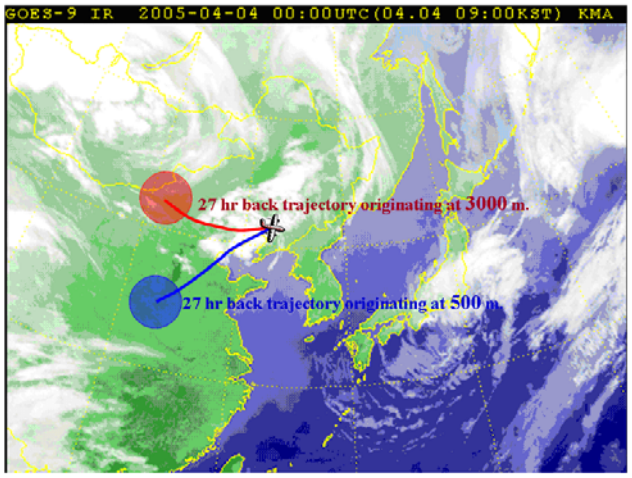

Figure 10. GOES9 IR image for 0000 UTC 4 April 2005, 27 h prior to the flight depicted in Figure 2. Back trajectories originating at 500 m (blue line) and 3000 m (red line) are superimposed; the transparent red and blue disks show the approximate area of origin for air sampled on 5 April. Note deep convective clouds to the W and NW and clear skies to the SW.

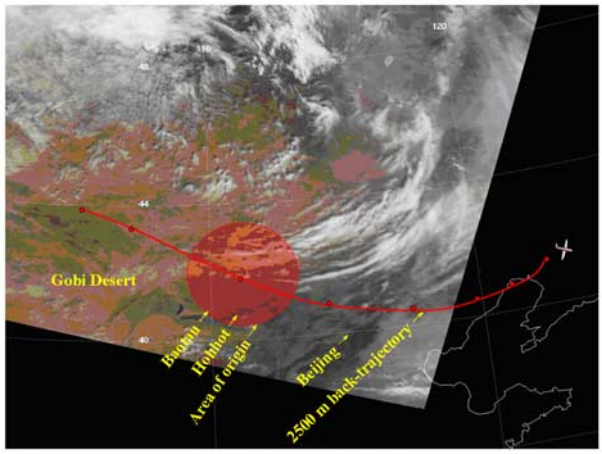

Figure 11. MODIS true color image for 0400 UTC 4 April 2005 with 2500 m back trajectory superimposed. Note heavy convection to NW over sparsely populated, arid regions of Mongolia and smaller bands of convective clouds intersecting the back trajectory. The transparent red disk indicates the approximate origin of the air sampled ~27 h later on the flight of 5 April. Baotou and Hohhot are industrial cities with substantial coal combustion and steel production.

confirmed that the in the warm sector where the low-level air originated, the sky was generally clear. Convection ahead of the cold front does not appear to have been the mechanism responsible for lofting in this event.

[19] Convective transport to the west (upstream at higher altitudes) of the flights would likewise not be captured by the back trajectories, but deep clouds could lift pollutants and dust (back trajectories cross the Gobi Desert) to high altitude where the westerlies would carry the trace gases and aerosols to the aircraft. Satellite images (Figure 10) show strong convection over arid NW Mongolia (in the top left corner of the image) and these clouds could have lofted dust to aircraft altitudes, but this part of the world is sparsely populated and is unlikely to be the source of the high concentrations of CO and SO_2 observed on 5 April. A smaller convective system is seen along the back trajectory (Figure 10) corresponding to the origin of the air 27 h prior to sampling. The MODIS IR cloud height analysis indicates that these cloud bands reached maximum heights of 6–12 km, and could have detrained pollutants and dust to flight altitudes. These clouds formed over a major steel producing region near the cities of Baotou and Hohhot, suggesting that this convective system was responsible for the widespread high concentrations of pollutants seen in the free troposphere ahead of the front (Figure 11).

4.2. Behind the Cold Front: 7 April 2005

[20] Following the flight of 5 April 2005, the cold front passed Shenyang; the dry, relatively warm continental tropical (cT) air mass moved off the coast and a cP air mass brought cooler, drier air (Figures 3 and 12). When a flight pattern similar to that of 5 April was carried out on 7 April in this cP air mass, the composition of the atmosphere was found to be substantially different (Figure 13). Pollutant levels were low with SO_2 mixing ratios well below 1 ppb and CO mixing ratios (not shown) below about 200 ppb. Particulate loading, however, was heavy with scattering coefficients ~8 × 10^{-5} m^{-1} corresponding to ~20 μg m^{-3} aerosol in the sub-μm mode at altitudes up to at least 4000 m. Larger particles may well have been present, but the inlet employed for this study is inefficient at sampling coarse mode aerosols. The NASA OMI Aerosol Index shows a broad maximum over Shenyang on 7 April 2005 (see web site given above). Ozone concentrations increased above the PBL, reaching a maximum of about 65 ppb,

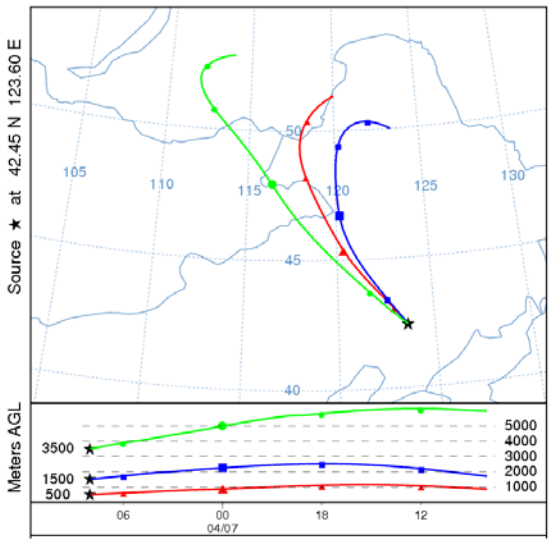

Figure 12. Back trajectories (36 h) calculated with HYSPLIT for air arriving at location of southern spiral (Figure 3). The plot at the bottom depicts vertical motions and shows subsidence in this postfrontal air.

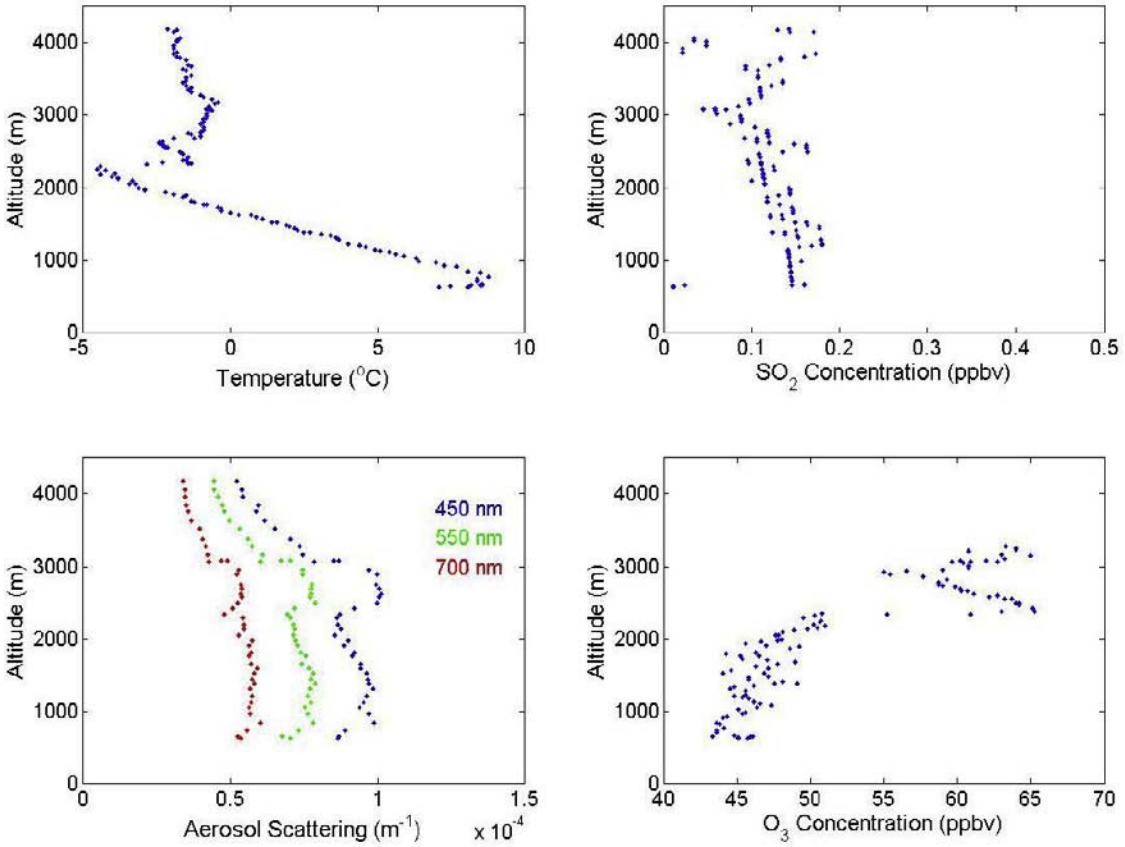

Figure 13. Profiles of trace gases and aerosol scattering for 7 April 2005 behind the cold front. SO_2 concentrations are low, but aerosol concentrations are high, approximately 25 μg m^{-3} sub-μm diameter particulate loading up 3000 m altitude and decreasing slowly aloft. Mineral dust from deserts to the NW is probably responsible for these aerosols. Ozone mixing ratios are consistent with downward transport from the UT/LS.

higher than expected for air with low concentrations of ozone precursors [e.g., *Logan*, 1999].

[21] Back trajectories (Figure 12) show rapid subsidence in the FT and brisk flow from the NW, an arid and sparsely populated region of China. The high wind speeds and low concentrations of pollution suggest that this was a dust event with surface material carried aloft on the turbulent winds from over the Gobi and other arid regions. Ozone was likely transported downward from the upper troposphere/lower stratosphere (UT/LS) rather than produced by local photochemistry.

[22] Full meteorological analysis for other flights including another frontal passage on 9–11 April where clouds and rain were seen ahead of the front will be discussed separately [*Li*, 2007]. A summary of results from all flights can be seen at the EAST-AIRE web site (http://www.atmos.umd.edu/~zli/EAST-AIRE/station.htm).

4.3. Observations of SO_2 With the OMI Satellite Instrument

[23] The OMI satellite instrument detected a major plume of SO_2 over NE China on 5 April 2005, but the column content derived from the first guess altitude profile was smaller than that obtained by integrating the aircraft-measured concentrations over altitude. When the air mass factor used in the OMI retrieval was corrected for the observed SO_2 profile shape, the derived column content more closely matched in situ observations (Figure 14). Altitude integrals of the SO_2 concentration measured from aircraft yield 1.3 DU over Xiaoming (42.45°N, 123.70°E) and 2.1 DU over Liaozhung (41.35°N, 122.65°E) near the midrange of OMI values. On the basis of the satellite observation, the total mass of SO_2 in the plume over NE China was estimated as 7.7×10^4 tons, with about 10% in the FT. As the cold front moved off the coast, the air quality over NE China improved dramatically.

[24] The cold front pushed the air mass containing high concentrations of SO_2 off the coast, the PBL air was caught in WCB circulation and lofted (Figure 15). Forward trajectory calculations (details available at http://www.atmos.umd.edu/~zli/EAST-AIRE/station.htm) indicate that the SO_2 plume ascended to between 2 and 4 km altitude. Using this new altitude in the retrieval, we derive about 5.7×10^4 tons of SO_2 in the event on 6 April 2005. By 7 April 2005 (Figure 16) the plume was over the North Pacific well east of Japan, and the mass of SO_2 had fallen to about 4.6×10^4 tons (Table 1). One day later, the plume had dissipated and the SO_2 in it is difficult to quantify.

[25] From the observed decay in SO_2 mass we can estimate its overall lifetime in this event. Assuming first-

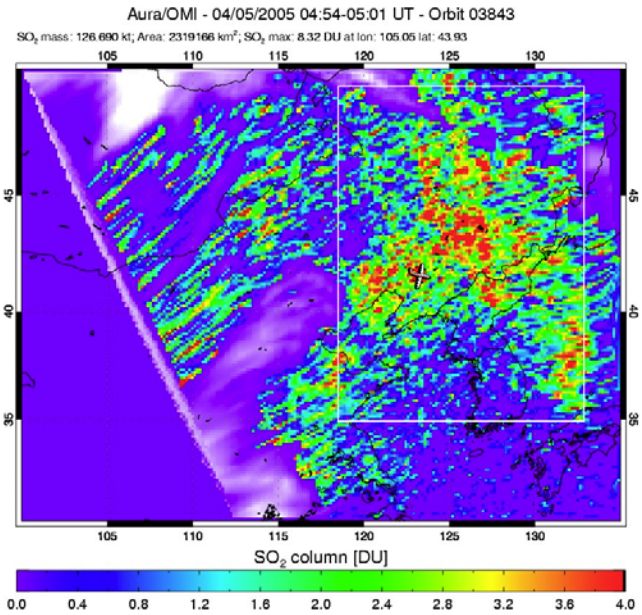

Figure 14. SO_2 column content (DU) derived from OMI observations for 5 April 2005, showing large mass of pollution over NE China. Axes depict longitude and latitude in degrees, and the aircraft symbol roughly shows the locations of the in situ profiles. The data were plotted with no cloud reflectivity filter for one orbit pass; pale white areas indicate clouds. The total mass of SO_2 calculated in the area outlined by the white box was 7.7×10^4 tons. The white diagonal line to the west results from the limit of the scan angle.

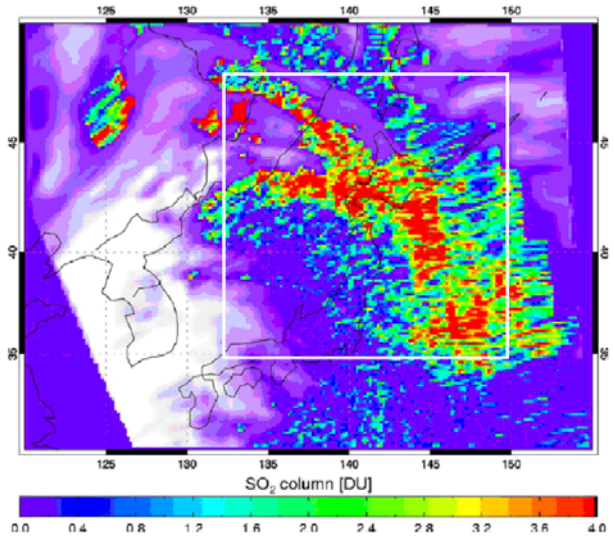

Figure 15. Same as Figure 14 but for 6 April 2005, showing large mass of pollution moving off the coast of NE China. The position of the SO_2 plume was estimated from four forward trajectories whose corners are outlined by the white box. The total mass of SO_2 estimated in the plume was 5.7×10^4 tons.

order loss, Figure 17 shows good linearity for the first three days; the points describe a line with a slope of about -0.26 d^{-1}. By 8 April the signal-to-noise ratio has diminished to the point of being unusable. We estimate the lifetime of SO_2 in this environment at 3–5 d, in the range of prior estimates [*Berglen et al.*, 2004; *Junkerman and Roedel*, 1983].

4.4. Comparison of China to North America

[26] While measurements made in a single season cannot form a chemical climatology, the results from the first series of flights of EAST-AIRE show that concentrations of trace gases and aerosols over China are substantially higher than are typically measured over North America. Over the past decade, aircraft measurements over the northeastern United States have amassed a substantial data set of trace gases and aerosol optical properties [*Prados et al.*, 1999; *Taubman et al.*, 2006; B. F. Taubman et al., Aircraft observations over the mid-Atlantic U.S. and North Atlantic Ocean during the 1996 Atmosphere/Ocean Chemistry Experiment: Evidence for linked transport of natural and anthropogenic ozone over Bermuda, manuscript in preparation, 2007, hereinafter referred to as Taubman et al., manuscript in preparation, 2007]. Figure 7 shows the mean of the CO mixing ratio measured on 445 flights made under smoggy conditions over the NE United States, the mean CO from two flights made in April 1996 ahead of a cold front over eastern North America, and CO data measured on four profiles flown ahead of a cold front approaching NE China on 5 April 2005. The concentration of CO over China was greater than that typically measured over the United States at all altitudes. Over the United States in summer, the 90th percentile CO mixing ratio ranges from 780 ppb near the surface to 280 ppb at 1000 m. Concentrations in the PBL were almost five times greater over China than over the United States for similar meteorological conditions. In spring time ahead of cold fronts, WCB lifting and convection increase concentrations of CO in the FT over both continents, but the great

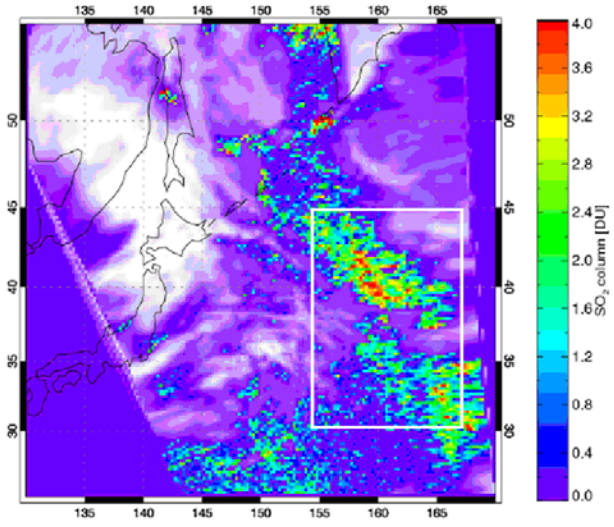

Figure 16. Same as Figure 14 but for 7 April 2005 when the plume of SO_2 was over the North Pacific. The position of the SO_2 plume was estimated from four forward trajectories outlined by the white box. The estimated total mass of SO_2 remaining was 4.6×10^4 tons.

Table 1. Properties of the SO_2 Plume as Measured by OMI for Figures 14–16[a]

Date of 2005	SO_2, ktons	Altitude, km
5 Apr	73–81	0–3
6 Apr	55–60	2–4
7 Apr	43–49	3–5

[a]The mass on 5 April 2005 was calculated using an AMF assuming all the SO_2 to be below the PBL. On the other three days, the mass was found assuming a Gaussian profile of SO_2 with a peak at the altitude derived form forward trajectory calculations. By 8 April the plume has become too diffuse to identify with reliability.

concentrations over China show the potential for a great global scale impact.

[27] The results for SO_2 and aerosol loading (Figure 8) also show greater concentrations over China. The SO_2 measured in the lower boundary layer of the United States [*Taubman et al.*, 2006] was a fraction of that measured over China. The boundary layer in summer is deeper than in spring and concentrations of SO_2 over China and the United States are comparable around 1000 m altitude, but above the boundary layer the mean SO_2 concentration over China is again sizable. For example, at 2500 m over the United States the mean mixing ratio of SO_2 is 0.2 ppb (with 90% of the data less than 0.6 ppb) and over China the mean is 0.6 ppb (90% less than 2.3 ppb). For aerosol scattering we can compare spring measurements over China to spring and summer measurements over the United States (Figure 8b). The scattering over china is greater than was measured under smoggy summer conditions over North America and an order of magnitude greater than was measured in the spring. At 2500 m for example, the mean b_{sp} over North America in summer is about 50 Mm^{-1} with 90% of the observations less than about 100 Mm^{-1}; the mean measured over China is about 250 Mm^{-1}. Full statistics for these flights can be found at the EAST-AIRE web site given above. These results are illustrative of the impact China may have on large scale atmospheric composition, but further flights are required to compile a chemical climatology for Asia.

5. Discussion

[28] Pollutants over China even if emitted in great amounts, have little impact on the global scale unless they are lofted above the PBL; rapid vertical transport of aerosols, NOx and SO_2 increases their lifetimes and range of influence. Both WCB lofting and convection play a role in transport of trace species into the FT. The cyclone described here generated little precipitation on 5 April, and over the continent vertical transport associated with it appears to be driven by dry convection well upwind. Dry convection, with minimal wet removal, may be especially effective at vertical transport of dust and pollutants.

[29] The new results presented here when combined with the climatology presented above may have general implications for climate and chemical transport models. Global-scale dynamical models can handle transport along isentropes, such as in the WCB explicitly; for the case study presented here this process was important only after the air mass was advected off the continent. Although existing climatologies do not address spring, the season of maximum pollution and dust transport, WCB processes can be quantified by dynamical models. Convection, in contrast, must be parameterized in large-scale models and accurate estimates of the aerosol and trace gas transport in dry convection poses a challenge to numerical models. Careful evaluation against observations and cloud resolving models is called for. Although EAST-AIRE has added to the database, the rate of emissions of pollution and dust in

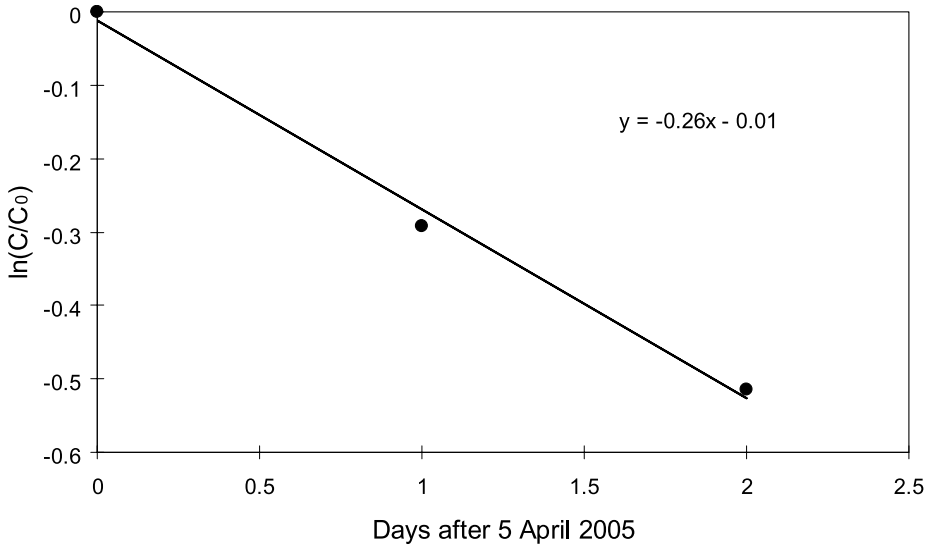

Figure 17. Decay plot for SO_2 over the Sea of Japan and North Pacific. Values for 6 and 7 April were calculated with an air mass factor (AMF) assuming the SO_2 distribution to be Gaussian around 3 km. The value on 5 April was calculated using an AMF assuming the SO_2 is below 3 km. Means from Table 1 were employed; the slope of the line gives an estimate of the first rate order removal rate of $\sim$0.26 d^{-1}; the implied lifetime is $\sim$4 d. The uncertainty is substantial; thus the implied lifetime is 3–5 d.

China and the fraction lofted remain major unanswered questions in global atmospheric chemistry and climate.

6. Conclusions

[30] Both dry convection and WCB lofting associated with wave cyclones can play an important role in the springtime interhemispheric transport of pollutants. The eastward propagation of extratropical cyclones and associated fronts provide the mechanism for lofting, removal, and long-range transport of dust and pollutants. Over the Asian continent in spring, dry convection appears to dominate, but as the systems move off the coast, WCB lofting becomes important. These mechanisms were investigated with satellite and research aircraft observations made over NE China in April 2005.

[31] In situ measurements showed that pollution concentrations in the PBL ahead of a cyclone and the associated cold front were several times higher than are typically found over eastern North America. Above the PBL (in the free troposphere at altitudes between ~1000 and 4000 m) concentrations of trace gases and aerosols were smaller, but much greater than background values; we measured ~300 ppb CO, ~2 ppb SO_2, ~70 ppb O_3, and ~8×10^{-5} m^{-1} aerosol scattering. Back trajectories show little or no WCB lifting over the continent, and we attribute the trace gases and aerosols found above the PBL to lofting by convective clouds that formed near the industrial areas of Hohhot and Baotou, near the boarder with Mongolia, on the previous day. The pollutants were carried in the westerlies to the aircraft. Convection was stronger to the northwest 48 h prior to the flights, but occurred over sparsely populated regions of Outer Mongolia. This area may have contributed dust to the FT but is unlikely to be the source of pollution; up until this point, dry convection appears to be the dominant mechanism of vertical pollutant transport.

[32] Behind the cold front, low concentrations of pollutants but high concentrations of aerosols (apparently mineral dust) and ozone were observed from the surface to the highest altitude flown, ~4000 m. High winds in the high-pressure area (cP air mass) can loft dust from the surface and subsidence or tropopause folding can bring ozone from the UT/LS to the middle troposphere.

[33] Satellite observations indicate that as the front moved off the coast, upward transport associated with flow along lines of constant entropy in the warm sector of the cyclonic system became an important mechanism in lofting of pollutants. The bulk of the SO_2 in the PBL, covering an area of roughly 2×10^6 km^2 and containing nearly 10^5 tons, was lifted into the LFT. This gas was observed to decay with an effective first-order rate constant of roughly 0.26 d^{-1} for a lifetime in the lower troposphere of 3–5 d. The WCB case study shown here lofted at least 5×10^4 tons of SO_2 enough to generate ~10^5 tons of ammonium sulfate aerosol.

[34] There are similarities in the meteorological mechanisms leading to long-range transport over eastern North America and over east Asia; both involve the passage of wave cyclones and associated fronts. However, one fundamental difference is that over NE China, little moisture is advected from the ocean onto the continent except in the warmest months, May to September. Convection over China in spring produces little rain; this makes convective lofting more efficient for pollution transport and makes convection harder to model. Future studies of the effects of emissions in China must account for dry convection; convective schemes of chemical transport models must be evaluated not only for their ability to match observed precipitation but also for their ability to simulate deep clouds that do not rain. Satellite observations of cloud tops and outgoing longwave radiation may be useful, but comparisons to observed tracers such as CO might be definitive. Further aircraft flights in the source regions will help quantify the role of dry convection and the WCB in large-scale atmospheric composition and climate.

[35] **Acknowledgments.** The EAST-AIRE project was supported by the National Science Foundation (ATM0412040), the NASA Radiation Science Program (NNG04GE79G), the National Science Foundation of China (40250120071), the Chinese Academy of Sciences (2003-2-9) and the Liaoning Provincial Meteorological Agency, CMPS of UMD, and the 973 National Basic Research Program of China (2006CB403706). We thank the Korean Meteorological Administration, especially S. W. Kim for weather information. R. Levy provided MODIS images. The authors dedicate this paper to Yoram Kaufman who was a friend, colleague, and inspiration to us all.

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