

Aircraft observations of dust and pollutants over NE China: Insight into the meteorological mechanisms of long-range transport

R. R. Dickerson, C. Li, Z. Li, L. T. Marufu, & J. W. Stehr
The University of Maryland
College Park, MD 20742
USA

H. Chen, P. Wang, X. Xia
Institute for Atmospheric Physics, Beijing
China

X. Ban, F. Gong, J. Yuan
Liaoning Meteorological Bureau, Shenyang, P.R. China

J. Yang
Nanjing University of Information Science and Technology, Jiangsu, P.R. China

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Abstract

Substantial concentrations of trace gases and aerosols are lofted and carried from Asia over the Pacific producing an inter-hemispheric impact on atmospheric chemistry and climate. The meteorological mechanism leading to this large-scale transport of dust and pollutants remains a major uncertainty in quantifying the global effects of emissions from the developing world. Models and downwind measurements have identified isentropic advection associated with wave cyclones (warm conveyor belt circulation) as an important mechanism. We present data from a case study conducted over Shenyang in NE China as part of EAST-AIRE in April 2005 in which upstream convection, rather than WCB lofting appears to dominate. Observations from instrumented aircraft flights, back trajectories, and satellite images of clouds (GOES) and aerosols (MODIS) are analyzed. In this heavily industrialized and populated region, the warm-sector PBL air ahead of a cold front was highly polluted. In the free troposphere, between ~1000 and 4000 m altitude, concentrations of trace gases and aerosols were lower, but well above background; we measured ~70 ppb O₃, ~300 ppb CO, ~2 ppb SO₂, and ~ 8x10⁻⁵ m⁻¹ aerosol scattering. These observations show that dry (non-precipitating) convection can be an important mechanism for converting local air pollution problems into regional or global atmospheric chemistry problems. Climatological data indicate that spring (MAM) precipitation over NE China is low, about 90 mm compared to 290 mm over the NE US. Cloud cover, however, is similar with cumulus clouds reported about 7% of the time over NE China and about 9% of the time over the NE US suggesting that lofting in dry convective events may be common over NE Asia. Evaluation of models' convective schemes and further observations near the source regions are called for.

Introduction

In the coming decades, much of the world's economic growth and associated atmospheric emissions will come from the developing world, with East Asia one of the major players, (e.g., [IPCC, 2001; Liu, *et al.*, 2005; Ma and van Aardenne, 2004; Simpson, 2006; Streets, *et al.*, 2003; Streets and Waldhoff, 2000; Tanre, *et al.*, 2005; Wang, *et al.*, 2005; Zhang, *et al.*, 2004]). Trace gases and aerosols emitted in China impact not only the local environment and human health, but appear to have an unusually broad range of influence. 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; Carmichael, *et al.*, 2003; Hannan, *et al.*, 2003; Holzer, *et al.*, 2005; Jaffe, *et al.*, 2003; Liang, *et al.*, 2004; Liu, *et al.*, 2003; 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 the N Atlantic and Europe, e.g. [Dickerson, *et al.*, 1995; Stohl, *et al.*, 2003] and Europe to Asia e.g., [Akimoto, 2003].

Key to determining the large-scale impact of emissions is quick transport out of the planetary boundary layer (PBL) into the free troposphere (FT). In the PBL, NO₂ and SO₂ 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 SO₂ is converted to sulfate aerosol that impacts atmospheric radiative balance and cloud properties. Two mechanisms, both associated with wave cyclones, may be involved [Cooper, *et al.*, 2004; Hess, 2005; Oshima, *et al.*, 2004]. Ahead of cold fronts, air rises along lines of constant entropy in what has been called warm conveyor belt circulation (WCB), and convection

causes rapid lofting of PBL air. Behind cold fronts, pollution levels tend to be low (see *Li et al.* this issue), 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].

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) [*Li*, 2006b]. We have developed a research platform for direct investigation of dust and pollutants in the source region. In this paper we present two 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 downwind.

Experimental Methods

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. Due to inlet line losses, sampling of super- μ m particles is inefficient, and measurements are representative of sub- μ m particles only. Temperature, relative humidity (RH), and pressure were measured using a thermistor, capacitive thin film, and Rosemount Model 2008 pressure transducer, respectively.

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) non-dispersive 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 SO₂. 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, currently be reanalyzed, will be presented elsewhere [Li, 2006a]. Aerosol scattering was determined with an integrating nephelometer (TSI Model 3563) that measured the total particle scattering coefficient (σ_{sp}) at 450, 550, and 700 nm [Anderson, *et al.*, 1996].

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 [Li *et al.* 2006], we flew a total of eight research missions on April 1, 5, 6, 7, 9, 10, 11, 12 under a variety weather conditions. 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 observations can be viewed from the EAST-AIRE homepage: http://www.atmos.umd.edu/~yuan/web_proj/air_camp/air_camp.htm

Flights described here include two spiral profiles in the northern and southern ends of the flight near Liaozhong and Tieling. At Liaozhong, a ground-based intensive observation station was installed to support of the air campaign measuring a large number of aerosol, radiation and cloud quantities [Xia *et al.* 2006]. Presented in this paper are results for two flights ahead and behind a cold front.

Results

Ahead of the cold front; 5 April 2005.

Substantial pollution was found aloft on 5 April 2005 when the aircraft flew ahead of a cold front (Figures 2-6). 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. Spirals were flown over rural areas to the north and south of the city. Concentrations of aerosols, CO, and SO₂ in the PBL were several times higher than are typically found over the eastern US during summer pollution events e.g., [Taubman, *et al.*, 2006]. Ozone mixing ratios reflect moderate photochemical smog formation, and show a peak just above the inversion and maximum in σ_{sp} (Figure 3) perhaps due to aerosol scattering of actinic radiation [Dickerson, *et al.*, 1997]. Concentrations of aerosols and trace gases fell off sharply above the strong (still 7 K at 11 am LST) inversion but remained well above background. These heavy loadings of pollutants and possibly mineral dust persisted to the highest altitudes sampled, ~4000 m. The aerosols detected on this flight were wide-spread 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 and MODIS page reference from Rob). These satellite observations show that this case study was an example of long-range transport of substantial concentrations of particulate matter.

The meteorological conditions for this flight (Figure 2) involved low-level flow from the south, through some of the most highly populated and polluted parts of China. This air from the warm sector ahead of the cold front brought heavy loadings of pollutants and dust (Figures 2-6) to the region around Shenyang. The system little

moisture or rain to the flight region until 6 April (flight discussed separately; [Li, 2006a]), but by 7 April only cirrus clouds were observed. In the area of the flights and to the south few clouds were observed.

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 7. Flow at low levels was from the SW while flow at higher altitudes originated from the W. Little evidence of upward motion is seen in the pre-frontal zone along the east coast of China, suggesting that the WCB was not effective in lofting pollutants 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 IR images for 3–5 April 2005 for clouds, and confirmed that 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.

Convective transport to the west (upstream at higher altitudes) of the flights would likewise not be captured by the back-trajectories, but 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 8) 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 CO and SO₂ observed on 5 April. A smaller convective system is seen along the back trajectory (Figure 8) 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.

Behind the cold front; 7 April 2005.

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 continental polar (cP) air mass brought cooler, drier air (Figures 10 & 11). 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 substantially different (Figure 12). Pollutant levels were low with SO₂ mixing ratios well below 1 ppb and CO mixing ratios (not shown) below about 200 ppb. Particulate loading, however, was heavy with scattering coefficients approaching 10⁻⁴ m⁻¹ corresponding to ~15 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 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.

Back-trajectories (Figure 11) show rapid subsidence 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.

Other flights including another frontal passage on 9-11 April where clouds and rain were seen ahead of the front and by the airplane – these will be discussed separately

[Li, 2006a]. Preliminary results from these experiments can be seen at the EAST-AIRE web site (http://www.atmos.umd.edu/~yuan/web_proj/station.htm) and will be detailed elsewhere.

Discussion

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, NO_x 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 vertical transport associated with it appears to be driven by dry convection well upwind. Here we examine whether this may be generally true for springtime aerosol transport events over China.

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 13). Over North America, cold fronts often demark the collision of maritime tropical (mT) and 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 US 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 US (Figure 13).

In contrast to precipitation, the total average cloud cover over NE China is not

dramatically different from that over the NE US. Over mid-latitudes (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 US. The peak in the diurnal cycle at both locations falls at 13 to 14 LST; cumulonimbus clouds are present in about 1% of the observations of both continents. Aerosols seem to have reduced cloud cover over China over the past few decades, but not enough to change the basic climatology [Qian, et al., 2006; USDOE, 1986]. In short, convective clouds form over China, but in spring little rain falls. Dry convection, with minimal wet removal, may be especially effective at vertical transport of dust and pollutants.

The results presented here, especially the identification of dry convection as a key meteorological mechanism, may have general implications for global climate and chemical transport models. Global-scale dynamical models can handle transport along isentropes, such as in the WCB explicitly; this process, although not important for the case study presented here, undoubtedly plays a role in many events and is probably well quantified by dynamical models. Convection, in contrast, must be parameterized 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 data base, the rate of emissions of pollution and dust in China and the fraction lofted in wet and dry convection remain major unanswered questions in global atmospheric chemistry and climate.

Conclusions

Dry convection associated with wave cyclones can play an important role in inter-hemispheric transport of pollutants. The eastward propagation of extra-tropical cyclones and associated fronts provide the mechanism for lofting, removal and long-range transport of dust and pollutants from local sources. This mechanism was investigated using data acquired from research aircraft flights over NE China in April 2005. This paper focuses on flights ahead of and behind a cold front.

Near a cyclone and the associated cold front, substantial pollutants and mineral dust were found aloft, but back trajectories show little or no WCB lifting. 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 border 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 free troposphere but is unlikely to be the source of pollution. Upward transport associated with flow along lines of constant entropy in the warm sector may be an important mechanism in lofting of pollutants, but for this case study, dry convection appears to have dominated.

Behind the cold front, low concentrations of pollutants but high concentrations of aerosols (apparently mineral dust) and ozone were observed from the surface to ~4000 m, the highest altitude flown. 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.

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. But 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 is modest and produces little rain. Future studies of the global 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 in large-scale atmospheric composition and climate.

Acknowledgments

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Figure Captions

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.

Figure 2. Surface analysis (Korean Meteorological Association) for 00UTC 5 April 2005 showing a low-pressure system (cyclone) moving towards 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. Note little cloud cover or precipitation was reported in the vicinity of the flights.

Figure 3. Altitude profile of ozone, RH, and temperature measured south of Shenyang at 11:00 local time on 5 April 2005. Note maximum in RH and ozone near the strong temperature inversion (7 K) near 1000 m altitude. In the lower free troposphere the air is dry and ozone concentrations are lower.

Figure 4. Altitude profile of aerosol scattering coefficient, σ_{sp} , for the morning (~1100 LST) flight of 5 April 2005. Strong scattering in PBL reflects the heavy aerosol loading (~100 $\mu\text{g m}^{-3}$ for a scattering efficiency of 4 m^2/g). Note rapid drop in scattering above the inversion (Figure 3) but the absolute value of σ_{sp} remains high. Integrating the σ_{sp} over altitude yields an aerosol optical depth near unity at 550 nm.

Figure 5. Altitude profile of carbon monoxide for all data collected on flight of 5 April 2005. Mixing ratios drop off sharply above the inversion near 1000 m, but remain well above the background. The mean CO profile measured on smoggy summer days over the NE US [Taubman, *et al.*, 2006] is shown for reference.

Figure 6. Altitude profile of sulfur dioxide for all data collected on flight of 5 April 2005 (triangles) and all eight flights conducted over China (diamonds). For the flights ahead of the cold front (5 April 2005) mixing ratios drop off sharply above the inversion near 1000 m, but remain well above the background. The mean SO₂ profile measured on smoggy days over the NE US [Taubman, *et al.*, 2006] is shown for reference.

Figure 7. Back trajectories calculated with HY-SPLIT 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). Lower panel depicts vertical motions and shows little evidence of WCB lofting in the pre-frontal zone for this episode. Back trajectories with end points at the second spiral, 300 km north and 2 h later (CAN LI please verify) are similar.

Figure 8. GOES9 IR image for 00 UTC 4 April 2005, 27 h prior to the flight depicted in Figure 2. Back trajectories originating at 500 m (red line) and 3000 m (blue 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 9. 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.

Figure 10. Surface analysis (Korean Meteorological Association) for 00 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.

Figure 11. Back trajectories calculated with HY-SPLIT for air arriving at location of southern spiral (Figure 10). Lower panel depicts vertical motions and shows subsidence in this postfrontal air.

Figure 12. Profiles of trace gases and aerosol scattering for 7 April 2005 behind the cold front. SO₂ concentrations are low, but aerosol concentrations are high – approximately 15 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.

Figure 13. Climatology of precipitation near Baltimore and Beijing [Arakawa, 1969; Bryson, 1969]. Note that most of the rainfall over NE China comes in the summer while over the NE US it is more evenly distributed. In the spring, frontal passage 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].

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