Atmos. Chem. Phys. Discuss., 14, 28019–28077, 2014 www.atmos-chem-phys-discuss.net/14/28019/2014/ doi:10.5194/acpd-14-28019-2014 © Author(s) 2014. CC Attribution 3.0 License.



This discussion paper is/has been under review for the journal Atmospheric Chemistry and Physics (ACP). Please refer to the corresponding final paper in ACP if available.

Uplifting of carbon monoxide from biomass burning and anthropogenic sources to the free troposphere in East Asia

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Received: 2 September 2014 - Accepted: 7 October 2014 - Published: 12 November 2014

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Published by Copernicus Publications on behalf of the European Geosciences Union.



Abstract

East Asia has experienced rapid development with increasing CO emission in the past decades. Therefore, uplifting CO from the boundary layer to the free troposphere in East Asia can have great implications on regional air quality. It can also influence global

- ⁵ climate due to the longer lifetime of CO at higher altitudes. In this study, three cases of high CO episodes in East Asia from 2003 to 2005 are examined with spaceborne Measurements Of Pollution In The Troposphere (MOPITT) data, in combination with aircraft measurements from the Measurement of Ozone and Water Vapor by Airbus In-Service Aircraft (MOZAIC) program. High CO abundances of 300–550 ppbv were observed in
- MOZAIC data in the free troposphere during these episodes. These are among the highest CO abundances documented at these altitudes. Correspondingly, elevated CO was shown in MOPITT daytime data in the middle to upper troposphere in the 2003 case, mostly in the lower to middle troposphere in the 2004 case, and in the upper troposphere in the 2005 case. Through analyses of the simulations from a chemical trans-
- ¹⁵ port model GEOS-Chem and a trajectory dispersion model FLEXPART, we found different CO signatures in the elevated CO and distinct transport pathways and mechanisms for these cases. In the 2003 case, CO from large forest fires near Lake Baikal dominated the elevated CO, which had been rapidly transported upward by a frontal system from the fire plumes. In the 2004 case, anthropogenic CO from the North China Plain
- experienced frontal lifting and mostly reached ~ 700 hPa near the East China Sea, while CO from biomass burning from Indochina experienced orographic lifting, leeside-trough induced convection, and frontal lifting through two separate transport pathways, leading to two distinct CO enhancements around 700 hPa and 300 hPa. In the 2005 case, high CO of ~ 300 ppbv, observed in the MOZAIC data around 350 hPa, origi-
- nated from the anthropogenic source over the vicinity of the Sichuan basin and biomass burning from Indochina, after convection and strong frontal lifting. These cases show that topography affects vertical transport of CO in East Asia via different ways, including orographic uplifting over the Hengduan Mountains, assisting frontal lifting in



the North China Plain, and facilitating convection in the Sichuan basin. In particular, topography-induced leeside troughs over Indochina lead to strong convection that assisted CO uplifting to the upper troposphere. This study shows that the new daytime MOPITT near-infrared (NIR) and thermal-infrared (TIR) data (version 5 or above) have enhanced vertical sensitivity and may help qualitative diagnosis of vertical transport processes in East Asia.

1 Introduction

Carbon monoxide (CO) plays several important roles in the atmosphere. The oxidizing capability, an ability of the atmosphere to cleanse itself, is strongly influenced by the

- ¹⁰ CO level in the troposphere. CO near the surface is a major pollutant, harmful to human health. Under high NO_x conditions, CO is a precursor of ozone, while in low NO_x airmasses, CO helps ozone destruction (Jacob, 1999; Holloway et al., 2000). As carbon dioxide (CO₂) is produced in both ozone production and destruction processes (Holloway et al., 2000), CO is linked to the global carbon cycle (Suntharalingam et al., 2004;
- Yurganov et al., 2008; Nassar et al., 2010) affecting climate change. With a lifetime of weeks to months, CO is a good tracer tracking transport of pollution. In the purview of these roles, it is important to understand processes influencing the CO distribution and variability in the atmosphere.

Although the main sources of atmospheric CO and its mean status are generally
understood (Novelli et al., 1998; Jacob, 1999; Holloway et al., 2000), many processes influencing CO variations at different time scales are not well known. Uplifting CO from the boundary layer to the free troposphere (FT) is such a process, which usually occurs on the synoptic scale that spans hundreds to thousands of kilometers in space and lasts hours to days in time (Daley, 1991). Uplifted CO usually has a longer lifetime and
can be transported fast by the upper layer winds over long distances through continents and between hemispheres in the troposphere (Stohl, 2001; Stohl et al., 2002; Damoah



processes (1) frontal lifting, (2) orographic lifting, and (3) deep convection (Brown et al., 1984; Banic et al., 1986; Dickerson et al., 1987; Bethan et al., 1998; Pickering et al., 1998; Chung et al., 1999; Donnell et al., 2001; Kowol-Santen et al., 2001; Cooper et al., 2002; Liu et al., 2003; Miyazaki et al., 2003; Chan et al.; 2004; Mari et al., 2004; Li
⁵ et al., 2005; Liu et al., 2006; Kar et al., 2008; Zhao et al., 2008; Ding et al., 2009; Randel et al., 2010; Chen et al., 2012).

East Asia has experienced rapid development with increasing CO emission in the past decades (Duncan et al., 2007). In addition to impacts on local air quality (Wang et al., 2010), continuing increase in CO emissions will lead to great impacts on regional air quality (Jaffe et al., 1999; Bertschi et al., 2004) and climate (Berntsen et al., 1999) of the world because of an expected upward trend in pollution outflow from the

10

- region. East Asia is characterized by its unique and complex meteorology, topography, and land covers. Vertical transport of CO can be modulated by one or more of these conditions or by their interactions. For example, the likelihood of when and where ex-
- ¹⁵ tratropical cyclones are active is closely linked to the locations and frequency of frontal uplifting. Two regions are identified by Chen et al. (1991): one in the East China Sea and the Sea of Japan, the other over the leeside of the Altai-Sayan. Wet and dry convections prevail in different seasons in northern China because of the distinct climatological pattern in precipitation there (Dickerson et al., 2007). The topography there
- ²⁰ also plays an important role in uplifting of CO alone and/or interplaying with frontal systems, aiding convection in mountainous regions (Liu et al., 2003; Ding et al., 2009). Recently, Lin et al. (2009) proposed a new mechanism that emphasizes the impact of topography-induced leeside troughs over Indochina on strong convection. A variety of land cover types in East Asia make CO sources there diversified. In highly popu-
- ²⁵ lated urban areas, such as those in the North China Plain, anthropogenic emissions are high. Large biomass burning, occurring in areas with abundant vegetation, can generate great amounts of CO for vertical transport when meteorological conditions become favorable. Two such areas are Southeast Asia and the boreal forested area in Russia (Wotawa et al., 2001; Schultz, 2002; Duncan et al., 2003). So far, our under-



standing of the impacts of these processes and their interactions on CO uplifting is still rather limited (Dickerson et al., 2007). The objectives of studying vertical transport of CO in East Asia are to better understand the vertical distribution of CO in the region, to advance the assessment of impacts of long-range transport of Asian CO on regions

downwind, and to help improve simulating this process in atmospheric models on the synoptic scale, eventually leading to more realistic chemical weather forecast in the future (Lawrence et al., 2003).

Due to lack of continuous measurements, most studies on CO in East Asia are based on observations from periodic field campaigns (Jacob et al., 2003; Tsutsumi et al.,

- ¹⁰ 2003; Li et al., 2007; Ding et al., 2009) or simulations by chemical transport models (Berntsen et al., 1999; Bey et al., 2001) or both (Liu et al., 2003). CO measurements from satellites provide unprecedented data revealing CO variations over East Asia. One of the instruments is the Measurements Of Pollution In The Troposphere (MOPITT) (Drummond, 1992; Drummond and Mond, 1996), which is onboard of the Terra satellite.
- ¹⁵ MOPITT provides data of CO total column and CO vertical profiles at several altitude levels, which are retrieved using a nonlinear optimal estimation method theoretically based on the observed radiances and their weighting functions, the a priori information, and the retrieval averaging kernels (Rogers, 2000; Deeter et al., 2003). As a result, the MOPITT retrieval at one level can be contaminated by CO at other levels and thus
- MOPITT's vertical sensitivity becomes a concern in earlier applications of MOPITT data (Jacob et al., 2003). Nevertheless, a few studies (Deeter et al., 2004; Kar et al., 2004, 2006, 2008; Liu et al., 2006) demonstrated MOPITT's vertical sensitivity to some extent. Furthermore, the MOPITT data in new versions that use both thermal infrared and near infrared radiances have offered enhanced vertical sensitivity (Worden et al., 2010;
- Deeter et al., 2012). Therefore, a detailed examination of MOPITT's vertical sensitivity in East Asia, especially for its ability in detecting vertical transport of high CO episodes, is desirable.

In this study, three cases of high CO episodes in East Asia from 2003 to 2005 are examined with MOPITT satellite data, in combination with aircraft measurements from



the Measurement of Ozone and Water Vapor by Airbus In-Service Aircraft (MOZAIC) program (Marenco et al., 1998) (see Sects. 2 and 4). The vertical transport mechanisms are analyzed with simulations from a trajectory dispersion model FLEXPART (Stohl et al., 2005) and a chemical transport model GEOS-Chem (Bey et al., 2001),
along with other meteorology data and satellite fire data (see Sects. 2 and 4). The vertical sensitivity of MOPITT is first evaluated with the coincident MOPITT and MOZAIC data from 2003 to 2005 (see Sect. 3) and further illustrated throughout the analysis of the high CO episodes (see Sect. 4). Discussion on the three cases is synthesized in Sect. 5 and the major conclusions are provided in Sect. 6.

10 2 Model and data

2.1 Satellite MOPITT CO data

MOPITT is the first space instrument that targets continuous measurements of tropospheric CO. MOPITT has been onboard of the Terra satellite launched in 1999, making scientific measurements since March 2000. Terra is flying in a sun synchronous polar orbit with an altitude of 705 km, crossing the equator at ~ 10:45 and 22:45 LT and making 14–15 daytime and nighttime overpasses each day. MOPITT uses a cross-track scanning method with a swath of 29 pixels, each of them being 22 km × 22 km. Therefore, with a swath of ~ 600 km, only about one third of the global area is covered in a day. Additionally, clouds can cause even more gaps in MOPITT daily data.
This makes it challenging to use MOPITT data for synoptic studies. It takes 3 days to

- Ihis makes it challenging to use MOPITT data for synoptic studies. It takes 3 days to achieve a near-complete global coverage and 16 days for a complete global coverage. MOPITT measures upwelling radiation in two narrow infrared spectral regions for CO retrieval: (1) a thermal-infrared (TIR) band near 4.7 μm that has strong carbon monoxide absorption and (2) a near-infrared (NIR) band near 2.3 μm that has weak
- ²⁵ CO absorption. MOPITT Version 5 retrieval products are significantly different from earlier products and offer three distinct products depending on application requirements.



One of them is a TIR/NIR "multispectral" product, which has enhanced sensitivity to CO in the lower-most troposphere (Worden et al., 2010; Deeter et al., 2012). Validations and evaluations of MOPITT data in various versions are documented in Emmons et al. (2004), Worden et al. (2010), and Deeter et al. (2012).

⁵ We first examined the MOPITT vertical sensitivity through comparison between coincident MOPITT and MOZAIC CO profiles. Advances of Version 5 (V5, a TIR/NIR "multispectral" product) from Version 4 (V4, a TIR-only product) data were assessed. Then, V5 data were used throughout the case studies, in which MOPITT data were gridded horizontally into 0.25° latitude × 0.25° longitude and at the MOPITT vertical resolution of 100 hPa from the surface to 100 hPa.

2.2 Aircraft MOZAIC CO data

The Measurement of Ozone and Water Vapor by Airbus In-Service Aircraft (MOZAIC) program was initiated in 1993 by European scientists, aircraft manufacturers, and airlines to collect experimental data (Marenco et al., 1998). MOZAIC consists of auto-¹⁵ matic and regular measurements of ozone, CO, and water vapor by several long range passenger airliners flying all over the world. The aim is to build a large database of measurements to allow studies of chemical and physical processes in the atmosphere. In comparing MOPITT with MOZAIC CO data, coincident MOPITT and MOZAIC data from 2003 to 2005 were screened within a radius of 1.5° and within a 4 h period. Each MOZAIC profile was smoothed by applying the MOPITT averaging kernels and the a priori profile for the co-located retrieved MOPITT profile to account for the bias introduced by the averaging kernels and the a priori. Therefore, the smoothed MOZAIC CO profile \hat{x}^{MOZAIC} is derived by (Rogers, 2000)

$$\hat{\boldsymbol{x}}^{\text{MOZAIC}} = \boldsymbol{x}_{a}^{\text{MOPITT}} + \boldsymbol{\mathsf{A}} \left(\boldsymbol{x}^{\text{MOZAIC}} - \boldsymbol{x}_{a}^{\text{MOPITT}} \right)$$
(1)

where $\mathbf{A} = \partial \hat{\mathbf{x}} / \partial \mathbf{x}$ is the MOPITT averaging kernel matrix which describes the sensitivity of the MOPITT CO estimate to the true profile of CO, $\mathbf{x}^{\text{MOZAIC}}$ is the MOZAIC CO 28026



profile, which has been mapped to the MOPITT pressure grid. The quantity x_a^{MOPITT} is the MOPITT a priori, which is based on CO simulations from the MOZART model (Emmons et al., 2004).

The MOZAIC measurements usually extend from the surface to ~ 250 hPa. When validating MOPITT data using Eq. (1), CO mixing ratios above 300 hPa was supplemented with CO from the GEOS-Chem chemical transport model (see Sect. 2.6) on the same location and day, similar to the treatments by Emmons et al. (2004) and Worden et al. (2010), who used the MOZART climatology simulations. Because CO above 250 hPa is lower than that in the middle and lower troposphere, the bias due to this treatment is expected to be low.

2.3 The NCEP FNL meteorological data

The Final (FNL) global tropospheric analyses are on 1° by 1° grids every 6 h (http: //rda.ucar.edu/datasets/ds083.2/). The data are generated from the Global Data Assimilation System (GDAS), which continuously collects observational data from the Global

¹⁵ Telecommunications System (GTS) and other sources. The analyses are available on the surface, at 26 levels from 1000 hPa to 10 hPa, the tropopause, the boundary layer, two subsurface levels, and a few others. Parameters include surface pressure, sea level pressure, geopotential height, temperature, sea surface temperature, potential temperature, relative humidity, precipitable water, *u* and *v* winds, and vertical motion.

20 2.4 The MODIS fire count data

The Moderate-resolution Imaging Spectroradiometer (MODIS) is a type of instruments which have been onboard of the Terra (EOS AM) satellite since 1999 and on the Aqua (EOS PM) satellite since 2002. The MODIS fire products include a validated daily global active fire product (MOD14 Terra and MYD14 Aqua) (Justice et al., 2002), generated using a global active fire detection algorithm that uses a multispectral contextual ap-





pixel fire detection (Giglio et al., 2003). The horizontal resolution is 1 km. The fire data are acquired from the Fire Information for Resource Management System (FIRMS) (Davies et al., 2009).

2.5 The FLEXPART trajectory model

- To diagnose the transport processes and trace CO sources, we used the FLEXPART model (Stohl et al., 2005), which is a Lagrangian Particle Dispersion Model developed at the Norwegian Institute for Air Research in the Department of Atmospheric and Climate Research. FLEXPART can be driven by meteorological input data generated from a variety of global and regional models. In this study, the simulations were driven by the NCEP FNL data. This model has been extensively validated (Stohl et al., 1998; Cristofanelli et al., 2003) and widely used in studies of the influence of various meteorological processes on pollution transport (Cooper et al., 2004, 2005, 2006; Hocking et al., 2007; Ding et al., 2009; Barret et al., 2011; He et al., 2011; Chen et al., 2012). In running FLEXPART, a large number of particles are released from defined locations
 15 (latitude, longitude, and altitude) at a time. Backward or forward trajectories of the par
 - ticles are recorded in latitude (°), longitude (°), and altitude (km) every hour.

2.6 The GEOS-Chem chemical transport model

GEOS-Chem is a global three dimensional chemical transport model (http:// geos-chem.org). The model contains detailed description of tropospheric O₃-NO_x hydrocarbon chemistry, including the radiative and heterogeneous effects of aerosols. It is driven by assimilated meteorological observations from the National Aeronautics and Space Administration (NASA) Goddard Earth Observing System (GEOS) from the Global Modeling and Assimilation Office (GMAO). In this study, GEOS-Chem version v9-1-3 was employed, which is driven by GEOS meteorology with temporal resolution
 of 6 h (3 h for surface meteorological variables), with a horizontal resolution of 2° lati-



28029

tude by 2.5° longitude and 47 vertical levels, including \sim 35 levels in the troposphere from 1000 to 100 hPa.

GEOS-Chem uses anthropogenic emissions from the Emissions Database for Global Atmospheric Research (EDGAR) global inventory (Olivier and Berdowski, 2001), which
 are updated with regional inventories, including the emission inventory in Asia (Streets et al., 2006; Zhang et al., 2009). The biomass burning emissions are from the Global Fire Emissions Data (GFEDv3) monthly inventories (van der Werf et al., 2010) and biogenic VOC emissions are taken from the Model of Emissions of Gases and Aerosols from Nature (MEGAN) global inventory. Emissions from other natural sources (e.g., lightning, volcanoes) are also included.

The model has been extensively evaluated and used in studies of atmospheric chemistry and pollution transport (Bey et al., 2001; Heald et al., 2003; Liu et al., 2003; Liu et al., 2006; Zhang et al., 2006; Jones et al., 2009; Nassar et al., 2009; Kopacz et al., 2010; Jiang et al., 2011). GEOS-Chem can generally describe CO variability in the troposphere but somewhat underestimate the observations in the northern mid-latitudes possibly due to biases in the CO inventory or numerical diffusion in the model or both (Heald et al., 2003; Duncan et al., 2007; Nassar et al., 2009; Kopacz et al., 2010).

3 Comparison between MOPITT and MOZAIC CO profiles

MOPITT's vertical sensitivity can be described in terms of the averaging kernels (see

- Eq. 1) and the Degree of Freedom for Signal (DFS). The averaging kernel matrix indicates the sensitivity of the MOPITT CO estimate to the true CO profile, with I (identity matrix) being the best, when true CO profiles are retrieved, and 0 being the worst, when MOPITT retrievals just take the a priori. In reality, the average kernel matrix is less than I, implying some contaminations of CO from other levels to the retrieved level as that the OC verticed structure contaminations of CO from other levels to the retrieved level as that the OC verticed structure contaminations of CO from other levels to the retrieved level level as that the OC verticed structure contaminations of CO from other levels to the retrieved level of the table.
- so that the CO vertical structure cannot be fully resolved. DFS gives the number of independent pieces of information available vertically in the measurements and it is the sum of the diagonal elements of the averaging kernel matrix (Rogers, 2000). Fig-



ure 1 shows a yearly mean of DFS for daytime and nighttime, respectively, in East Asia for the V5 TIR/NIR data, indicating substantial increases in DFS compared to earlier MOPITT versions (Worden et al., 2010; Deeter et al., 2012). The daytime DFS in East Asia (Fig. 1a) ranges from 0.5 to 2.7, usually decreasing with latitude, similar to its distribution in other regions and on the global scale (Deeter et al., 2004; Worden et al., 2010). In the same latitudinal zones, the DFS is higher over land than over ocean. The daytime annual DFS is high in the Sichuan basin, the eastern part of mainland China, the Indochina peninsula, and the Indian subcontinent. Over the mountain or valley regions, DFS is low, such as above the Tibetan Plateau. The stars indicate the cities where MOZAIC vertical measurements are available for validation of MOPITT data.

- where MOZAIC vertical measurements are available for validation of MOPTTT data. The annual mean DFS is 1.65, 1.51, 1.60, and 1.64, respectively, in an area of 1° × 1° around Beijing, Narita, Shanghai, and Hong Kong, with a maximum of 1.98, 1.64, 1.81, and 1.74 for the cities, respectively. The nighttime DFS values (Fig. 1b) are lower (from 0.5 to 1.5) than the daytime values, similar to that in Deeter et al. (2004) for an earlier
 MOPITT version. Spatially, nighttime DFS is high over regions where the daytime DFS
- is also high.

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The general patterns of MOPITT averaging kernels have been documented (Pan et al., 1998; Edwards et al., 1999; Emmons et al., 2004; Deeter et al., 2003, 2004, 2012; Kar et al., 2008; Worden et al., 2010). For V5 MOPITT data, the averaging kernels at the four cities are similar to these in Worden et al. (2010, in their Fig. 7). The difference in the averaging kernels between V4 and V5 can be as large as 0.14 for the surface and lower troposphere and as 0.10 for the upper troposphere (not shown).

Figure 2 shows the relative bias between MOPITT and the smoothed MOZAIC $(\hat{x}^{\text{MOZAIC}})$ profiles (see Eq. 1), which is also referred as "MOPITT estimate of in situ" in Worden et al. (2010) and "transferred profile" in Emmons et al. (2004). For V5 data (in red), the mean bias is within ±20% for all the cities. In all the altitude levels, the bias is smallest (close to zero) around 500–400 hPa and increases upward and downward. The bias is mostly positive above 500–400 hPa, while below 500–400 hPa, it is



positive at Beijing, Narita but negative at Shanghai and Hong Kong. Whether the sign

change is related to the change in the geographic location (Shanghai and Hong Kong are both coast cities) can be a subject for further study. The V4 data (in green) also show the smallest bias in the middle troposphere. In the lower troposphere, the bias in V5 is reduced by 5–10% at Beijing and Narita. At Shanghai and Hong Kong, the bias
⁵ changes from positive in V4 to negative in V5, with a smaller (at Shanghai) or larger (at Hong Kong) magnitude. In the upper troposphere above 500–400 hPa, the bias in V5 at Beijing, Narita, and Shanghai changes to positive, with a magnitude as the same as or larger than in that in V4. At Hong Kong, the bias in V5 remains positive but the magnitude is enlarged. As a comparison, we also validated MOPITT data in other cities
¹⁰ in the globe and found that the mean bias in Europe or the United States is lower than in East Asia, especially in the surface layer (not shown).

The correlation between MOPITT and smoothed MOZAIC data is shown in Fig. 3 for the middle to upper troposphere (Fig. 3a) and for the surface to the middle troposphere (Fig. 3b). From 500 to 100 hPa, the correlation coefficient between the two data sets is 0.92, 0.86, 0.83, 0.68 at Beijing, Narita, Shanghai, and Hong Kong, re-

spectively (Fig. 3a), while from the surface to 600 hPa, the correlation becomes closer, being 0.90,0.92, 0.92, 0.94 at Beijing, Narita, Shanghai, and Hong Kong, respectively (Fig. 3b). The correlation coefficient between the two data is the best in the middle troposphere (500–400 hPa, not shown).

20 4 Uplifting of CO to the free troposphere

Daily MOPITT and MOZAIC data from 2003–2005 were screened to find cases of high CO episodes observed by both MOPITT and MOZAIC at the same location and time. We found three cases of high CO in MOPITT data with close-by MOZAIC measurements, while it was hard to find such high CO episodes with exact coincident MOPITT and MOZAIC observations because of large gaps in MOPITT data and limited aircraft sampling coverages. In the following, we provide detailed analyses of each case, ordered by year of occurrence in Table 1. The cases occurred over the East China Sea



or the Sea of Japan or both. High CO was shown in MOPITT daytime data in the middle to upper troposphere in case 2003, mostly in the lower to middle troposphere in case 2004, and in the upper troposphere in case 2005. For comparison, MOPITT and MOZAIC observations for the three cases are shown in Figs. 4–6, followed by analyses for each case with FLEXPART and GEOS-Chem simulations, in combination with MODIS fire data and NCEP FNL meteorological data. Table 1 provides a brief summary for all the cases. The cases occurred in spring and summer when cyclone activities are strong in East Asia (Chen et al., 1991; Yue and Wang, 2008). High CO concentrations up to 300–500 ppbv are observed in FT from 750 hPa up to 350 hPa. The CO sources are identified as biomass burning or a combination of biomass burning and anthropogenic origins. The outflow of the high CO episodes finally reached the boundary

4.1 Case study I: 6 June 2003

layer at the west coast of the United States and Canada.

On 6 June 2003, a large area (~ 400 km × 1500 km) of high CO up to 350 ppbv appeared in the MOPITT image over the Sea of Japan and the nearby continent in the middle to high troposphere (Fig. 4a). The area with high CO is the largest among the three cases, reflecting the strongest CO source from large forest fires in this case. In Fig. 5a, the MOPITT CO profile averaged over the boxed area in Fig. 4a shows a broad enhancement from the monthly profile between 800–300 hPa, with peak CO abundances

- of ~ 300 ppbv appearing around 550 hPa. The large difference between the MOPITT a priori and the measurements over these altitudes indicates MOPITT's capability of capturing pollution episodes with some degree of vertical sensitivity. This CO peak was not shown in the MOPITT monthly mean profile, reflecting the episodic nature of this event. The high CO episode was also captured by a near-by MOZAIC measure-
- ²⁵ ment (Fig. 5b). A layer of elevated CO is apparent between 500–350 hPa, with a CO peak up to ~ 550 ppbv around 400 hPa. In addition, the MOZAIC relative humidity (RH) and ozone profiles are shown in Fig. 5b. Around the altitudes of CO buildup, elevated humidity followed the CO profile, while ozone also showed some enhancement.



A latitude–altitude cross section from MOPITT is shown in Fig. 6a. It is the average between two blue dashed lines in Fig. 4a. The arrows represent the winds in the meridional and vertical directions and the contour represents the zonal wind speed. Consistent with Fig. 4a, high CO up to 350 ppbv appears in the middle to upper troposphere between 35–50° N. The CO buildup was with the highest abundance and the widest depth among the three cases, reflecting again the strongest CO source in this case.

To trace down the CO source, backward trajectories of the air particles were simulated using FLEXPART after releasing 30 000 and 7000 particles, respectively, from the locations of the large and small boxed areas in Fig. 6a (the same as the blue bars in Fig. 4a) on 6 June 2003 when CO was high in the MOPITT data. Because CO has a relatively long lifetime (weeks to month), it is assumed that CO is not removed in the backward trajectories. Figure 7 shows the distribution of particle concentration between 6.25–10.25 km (~ 500–250 hPa, Fig. 7a) and between 0–3.25 km (~ 1000–650 hPa,

- ¹⁵ Fig. 7b). The contour lines indicate the geopotential height at 850 hPa at 12:00 UTC on 3 June 2003 (Fig. 7a) and at 0 UTC on 2 June 2003 (Fig. 7b), respectively. The stars, diamonds, and circles in Fig. 7b show the location of large forest fires near Lake Baikal from MODIS fire data. The circles, diamonds, and stars denote daily mean fire counts of 20–100, 100–300, and 300–500 per 2.5° × 2.5° grid area, respectively, from 31 May
- to 6 June. The high particle counts between 0–3 km in the vicinity of Lake Baikal match well with the location of fire counts (Fig. 7b). On 3 June 2013, there was a cyclone with a cold frontal system (Fig. 7a) that rapidly lifted the CO originated from the fires along the warm conveyer belt (WCB) to the upper level. The particle distribution in the upper troposphere shows the transport pathway of the particles to the Sea of Japan.
- To further illustrate this, particles were released from the fire region near Lake Baikal (93–115° E, 50–65° N, 0–3 km, following Lavoue et al. (2000), who found an average injection height of Siberian fires of ~ 3 km). Forward trajectories were simulated and the resultant vertical distribution of the particles varying with time during 1–15 June 2003 is shown in Fig. 8. It is found that the released particles from the fires traveled along



the isobars to northeast of Lake Baikal from 1 June to 3 June 2003 and then the particles were lifted to the upper layers (3-8 km) on 3 June at 12 a.m. (in 60–70 h) (Fig. 8). Then, the particles were transported to the east along these altitudes. On 6 June (in 120–140 h), a large amount of particles appeared in a layer of 3-8 km (Figs. 8 and 4a).

⁵ The altitudes with high particle concentrations agree remarkably well the MOPITT data between 500–350 hPa (Figs. 4a and 6a).

It is the cyclone with a front northeast of Lake Baikal that transported the CO up along the WCB (Figs. 7a and 8). Figure 5b shows that the relative humidity reached about 65% in MOZAIC measurement, suggesting the air mass indeed came from a WCB

- (Cooper et al., 2002). The MOZAIC ozone profile also shows elevated ozone at the same altitudes but the shape does not follow exactly the ones of CO and humidity, implying complexity of chemical processes involved. The polluted air reached as high as 9 km although most particles remained at heights of about 3–8 km (Fig. 8). After being lifted to higher altitudes, the polluted air was transported by strong westerlies over long
- ¹⁵ distances. Figure 8 shows that the particles were further transported to the east and sink slowly after 7 June. Around 14 June 2003, the particles reached the east coast of Canada (0–5 km). The satellite MODIS data show a large number of hot spots near Lake Baikal in May and June 2003. Earlier studies have shown that forest fires in Asia can impact air quality in North America (Jaffe et al., 2004; Liang et al., 2004; Oltmans
- et al., 2010). This case illustrates again the role that WCBs played in the intercontinental transport of pollution for such high CO. It should be noted that the FLEXPART simulation was made by using the FNL meteorological data, which may have not considered the buoyancy force due to fires. Such buoyancy force can lift CO plumes even faster and higher. Our analyses are also consistent with Nedelec et al. (2005), who
- ²⁵ analyzed 320 MOZAIC flight routes from Europe to Asia in 2003 and reported the observations of high CO up to 800 ppbv above 8 km (~ 350 hPa) on 3 and 4 June 2003 around 57° N (northeast of Lake Baikal). This matches well with the time and location of frontal lifting of CO in this FLEXPART simulation. With analyses of different data sets, i.e., Along Track Scanning Radiometer (ATSR) fire data, the Total Ozone Mapping



Spectrometer (TOMS) aerosols data and the MODIS cloud data, Nedelec et al. (2005) also attribute the high CO at these altitudes to front lifting of CO from large forest fires near Lake Baikal.

4.2 Case study II: 18 March 2004

- ⁵ This case occurred on 18 March 2004 when high CO appeared in the MOPITT data in the lower and middle troposphere over the East China Sea (Fig. 4b). The elevated CO of 200–250 ppbv is observed between 750 hPa and 550 hPa vertically in MOPITT data (Fig. 5c). The departure of the MOPITT CO profile from its a priori reflects the MOPITT's vertical sensitivity (Fig. 5c). The MOPITT monthly mean, like for the other two cases, follows a typical CO profile pattern with CO concentrations being the highest near the surface and decreasing gradually with altitude. The CO on 18 March 2004 was ~ 50 ppbv higher than the monthly mean above 800 hPa. A layer of large elevated CO appeared in the MOZAIC profile between 750–550 hPa with a peak of 500 ppbv around 650 hPa (Fig. 5d). The high RH (~ 90–100 %) below 600 hPa in the MOZAIC
- ¹⁵ data suggests that the air mass experienced some uplifting process that enhanced its humidity, likely from a WCB. The MOZAIC ozone peaked (~ 70 ppbv) around the same altitudes as CO, implying that ozone may be produced in the air mass carrying high CO during the transport process. Figure 6b shows a latitude–altitude cross section averaged between the two blue dashed lines in Fig. 4b. Around 30° N, elevated CO levels
- (~ 200 ppbv) are evident around 700 hPa. The corresponding winds (blue lines and arrows) show that there were strong descents north of the elevated CO and moderate ascents in the south of the region (Fig. 6b), indicating the formation of a frontal system with downward and upward flows, south and north of the front, respectively.

This case was simulated with GEOS-Chem to identify the sources of CO and to explore the transport mechanisms. The MODIS fire data suggest biomass burning over northern Indochina peninsula to be a source for the observed high CO (Fig. 9). The time series of fire counts over area of 20–25° N and 92–105° E peaked on 12 March 2004. Correspondingly, high CO of ~ 300 ppbv appeared in the MOPITT composite of



11–18 May 2004 at 700 hPa over northern Indochina peninsula (Fig. 9). This source is confirmed in the GEOS-Chem simulation (Fig. 10b). In addition, the anthropogenic source concentrated over the North China Plain (NCP) (approximately $30-40^{\circ}$ N, 110° – 125° E) is identified as another source of high CO in the GEOS-Chem simulation

⁵ (Fig. 10c). Figure 11 shows the latitude–altitude cross sections of the GEOS-Chem simulations of CO, fire-induced CO, and anthropogenic CO, respectively, along 130° E on 18 March 2004. The CO from biomass burning was more widely spread to the south than the anthropogenic CO. CO abundances from both sources were high around 700 hPa (Fig. 11b and c) between 25–35° N across 130° E where MOPITT observed high CO (Fig. 6b).

The different CO distributions for the two sources in three dimensions (Figs. 10 and 11) reflect rather different transport pathways and uplifting mechanisms. The transport of the fire-induced CO can be divided into the following four processes. First, the CO was orographically lifted along the Hengduan Mountains from the surface to \sim 750 hPa.

- ¹⁵ The lifted CO is shown in Fig. 12 around 100° E on a longitude-altitude cross section along 22° N. Then, the CO experienced two separate transport pathways. In the second process, part of the lifted CO was further transported upward to 400–300 hPa, shown as a bulb in Fig. 12 around 105° E. This is due to strong convection, possibly caused by by a frontal system developed on March 17, 2004(Fig. 9), interplayed with the leeside
- troughs east of the Hengduan Mountains. The vertical velocity reached 0.2 m s⁻¹ in FNL data around at this level (not shown). The ECMWF data also show northeastward airflow from Indochina peninsula with high potential energy (warm and wet) available for strong convection. All of these suggest that the strong convection over the leeside troughs rapidly lifted CO up to ~ 350 hPa. In fact, the orographic lifting and topography-
- induced convection are quite common in this region so high CO often appears at these two altitudinal levels in March as simulated by GEOS-Chem (not shown). On 17 March, the CO concentrations were even higher (~ 500 ppbv) around 400 hPa than the monthly mean because of the high CO source from large forest fires. In the third process, the uplifted CO around 400–300 hPa (near 105° E in Fig. 12) was transported northeastward



by strong winds along the front in the upper troposphere, reaching the East China Sea (near 30° N, 130° E) on 18 March (Figs. 10b and 11b). This transport enables high CO from forest fires in southern Asia in low latitudes to rapidly reach the upper troposphere in the mid-latitudes. In the fourth process, paralleling to the second and third, part of the

- orographically lifted CO stayed around ~ 700 hPa because of leeside-trough induced convection. This CO was transported eastward along the isobars of the low pressure system around 700 hPa (Figs. 10 and 12). This process occurred at lower altitudes than processes two and three. The transport was slower and it took longer time (from 15 to 18 March) for the CO to reach the East China Sea. Processes two and three brought
- CO to the upper troposphere, while process four increased CO in the lower to middle troposphere (Fig. 11b). For the anthropogenic CO in the North China Plain, the vertical transport was mainly carried out by frontal lifting. Horizontally, anthropogenic CO was transported eastward along 30° N (Fig. 10c). Consequently, the total CO shows a buildup centered near 700 hPa around 30° N and 130° E, mostly coming from the two CO sources (Fig. 11a, a).
- ¹⁵ CO sources (Fig. 11a–c).

Comparison of the simulated vertical structure of CO (Fig. 11a) with the MOPITT observation (Fig. 6b) shows that MOPITT can generally capture vertical transport of CO from forest fires and anthropogenic sources, although the magnitude of CO in MOPITT data was lower and there were also substantial gaps in the MOPITT images

- ²⁰ due to convective clouds. In the MOPITT data, high CO of ~ 200 ppbv reached up to 200 hPa. In the lower to middle troposphere, elevated CO (~ 200 ppbv) was centered around 650–700 hPa. These features are similar to the GEOS-Chem simulations. Note that the CO buildup around 300 hPa in the GEOS-Chem simulation (Fig. 11a and b) was reflected in the MOPITT data (Fig. 6b), but not as obvious as in the simulation
- since the MOPITT retrievals are smoothed with the averaging kernels. This CO is also shown as a little bump around 300 hPa in MOPITT vertical profile in Fig. 5c. This buildup is missing in the MOZAIC profile (Fig. 5d) because the aircraft flew towards the north and outside the region with high CO.



In FLEXPART, air particles were released over the large fire regions from the surface to 1 km on 11 March 2004. FLEXPART was used to simulate transport of the air parcels until 18 May 2004 at 2 a.m. Taking the same zonal means as for Fig. 6b, it is found that the vertical distribution of particle concentrations is similar to that in Fig. 6b with highest particle concentrations between 4–5 km (not show). As simulated by FLEXPART, the

outflow of the high CO finally reached the west coast of the United States with particles mainly distributed around 5 km in altitude. High CO observed in East Asia in this case appeared the most southerly among the three, leading to a most southerly outflow.

The transport mechanism of biomass burning by the fourth processes was first proposed by Lin et al. (2009) who found a significant role that the leeside troughs above the Indochina peninsula play in uplifting of ozone from biomass burning as there is lack of tropical deep convection to explain such strong convection. It is the interplay of the leeside troughs and the cyclone in the northeast of China which formed a front system that transported CO from the Indochina peninsula upward.

15 4.3 Case Study III: 10 April 2005

In this case, MOPITT observed high CO of ~ 250 ppbv at 300 hPa near the east coast of Japan on 10 April 2005 (Fig. 4c). To compare MOPITT and MOZAIC data, the mean MOPITT profile was taken over a boxed area (in Fig. 4c) upwind of the MOZAIC measurement. The MOPITT vertical profile clearly shows a CO peak around 300 hPa, where it departs from the MOPITT monthly mean (Fig. 5e). Comparing with cases 2003

- 20 where it departs from the MOPTTT monthly mean (Fig. 5e). Comparing with cases 2003 and 2004, MOPITT CO peaked at higher altitudes, illustrating some MOPITT vertical sensitivity even at these altitudes. In Fig. 5f, a sharp peak of 300 ppbv in MOZAIC CO is shown around 350 hPa. The profile of relative humidity followed closely that of CO, with values up to 90–100 % around 300 hPa, implying that the elevated CO was lifted
- to this level from the lower troposphere by a cyclone system along its WCB. However, the MOZAIC ozone profile varies strongly from the CO and relative humidity profiles. This was found to be due to a strong stratospheric intrusion introduced by the cyclone. HYSPLIT simulations suggest that a large amount of airmass plunged around 4 June



from 9 to 3–4 km over northwest of China, bringing high ozone to the lower troposphere (not shown). Miyazaki et al. (2003) also observed downwelling of stratospheric air on the back side of cyclones. Figure 6c shows an altitude–latitude cross section averaged between 120–150° E (between two dashed lines in Fig. 4c). High CO of 200–250 ppbv appeared between 300–200 hPa around 35° N. This is a rare case in which MOPITT

appeared between 300–200 nPa around 35 N. This is a rare case in which MOPTT1 reports such high CO (200–250 ppbv) at these high altitudes (around 300 hPa). Documented CO abundances observed by MOPITT at these altitudes were ~ 130 ppbv over Indian summer monsoon seasons (Kar et al., 2004), 110–150 ppbv in the North America from the forest fires, chemical, and anthropogenic sources (Liu et al., 2005; 2006), and ~150 ppbv in spring at Hong Kong (Zhou et al., 2013).

The MODIS fire data show that there were indeed large fires over Indochina peninsula in 3–10 April 2005, shown as stars in Fig. 13. Using GEOS-Chem, CO from fire and anthropogenic sources was simulated to identify their respective contributions and transport pathways.

- The entire process of vertical transport of CO is well reproduced by GEOS-Chem (Fig. 14). Figure 14a provides the CO distribution in the lower troposphere on 8 April 2005, while Fig. 14b and c show the CO distribution on the next day and the day after in the middle and upper troposphere, respectively. The geopotential heights at 750, 450, and 250 hPa are overlaid with the CO image for each layer accordingly. On 8 April
- 2005, there was a cyclone developing in the east of Lake Baikal between 110–120° E, 45–55° N. The surface CO was transported upward and northeastward along the WCB (Fig. 14a). On 9 April, the cyclone moved to the east (Figs. 13 and 14b). The high CO shows a "comma" shape along WCB at the mid-troposphere; this shape is typical for a mature cyclone system with a WCB (Cooper et al., 2002). The cyclone further
- ²⁵ moved eastward and reached the Sea of Japan on 10 April (Fig. 14c). The GEOS-Chem simulation shows accumulation of high CO over the ridge of high pressure and along the front at the upper troposphere. The GEOS-Chem simulations suggest that the outflow of the high CO reached Canada on 16 April.



The combined effects of cyclone activities, topography, and CO from different sources and locations are reflected in distinct CO signatures along the WCB. Figure 15 shows the CO from the fires (Fig. 15a and c) and from the anthropogenic source (Fig. 15b and d) in the middle and upper troposphere, respectively, overlaid with the geopotential height at 450 hPa (Fig. 15a and b) and 250 hPa (Fig. 15c and d), respectively. In the middle troposphere (500–400 hPa), a large amount of CO from the fires in Indochina peninsula was uplifted to this level along the middle part of the WCB on 9 April 2005 and was transported eastward on 10 April, 2005 (Fig. 15a). One source of the anthropogenic CO was concentrated around the North China Plain (Ding et al., 2009) where high CO was evident in MOPITT data (Fig. 13, 35–45° N, 100–120° E) (Fig. 15b). On 8 April, this CO was uplifted along the WCB and further transported to the middle troposphere, coming across sudden elevated terrains on the way and forming the head of the "comma" in the cyclone system (Figs. 14b and 15b). The topog-

the North China Plain associated with elevated terrain, and by Ding et al. (2009), who speculated possible topography lifting in North China Plain. In the southern end of the WCB (near 30° N, 120° E in Fig. 15b), the CO came from the anthropogenic source in the vicinity of the Sichuan basin (~ 26–34° N, 102–110° E). This CO was transported vertically to 500 hPa on 8 April at 18:00 UTC to 9 April at 00:00 UTC. Air pollution often accumulates in the Sichuan basin because of its special topography. The development

raphy's role was noticed by Liu et al. (2003), who found a ring of convergence around

accumulates in the Sichuan basin because of its special topography. The development of small scale cyclones there is well known as the southwest vortex or Sichuan low (Tao and Ding, 1981). Accumulated pollutants there usually are transported to the free troposphere by such convection. The strong convection can last more than 6 h and peak at the midnight (Yu et al., 2007). As this anthropogenic source is quite stable, its
 contribution should not be understated.

Interestingly, Lin et al. (2009) reported an observed ozone enhancement from ozonesonde data at 4 km in Taiwan on 11 April 2005. They proposed a new transport mechanism from their study as discussed in Sect. 4.2, in which they attributed the elevated ozone to the biomass burning in Indochina. Similarly, CO from biomass



burning is also apparent over Taiwan at the midtroposphere in the GEOS-Chem simulation (Fig. 15a), although the maximum CO enhancement was north of Taiwan at this altitude.

- The white dot in Fig. 15 indicates the location where MOZAIC passed over. It is clear that MOZAIC measurement was within the WCB at 200–300 hPa, while it was at a distance from the WCB at 500–400 hPa. This is consistent with the MOZAIC CO profiles shown in Fig. 5f, suggesting that MOZAIC in fact measured air from the stratosphere at these altitudes. As the wind was stronger in the upper than in the lower troposphere, the WCB-transported CO reached further east in the upper levels (Fig. 15). The simulations
- ¹⁰ suggest that over the boxed area in the MOPITT image in Fig. 4c at 300 hPa, the fire and anthropogenic sources each contributed about ~ 20 % of the observed CO. Comparing the GEOS-Chem simulation (Fig. 14c) with the MOPITT observation (Fig. 4c), we noticed that there were large gaps in MOPITT data north of 35° N where CO abundances were even higher than the MOPITT measurements south of 35° N. These gaps
- ¹⁵ were caused by clouds associated with the cyclone system. The complication due to clouds is a problem with an optical instrument like MOPITT, which explains why it is hard to find cases like this in which high CO can be observed by both MOPITT and MOZAIC under a frontal condition.

In this case, the strong part of the front (close to the centre of the cyclones) swept southern China, where CO was high (Fig. 13). Along the front $(30-40^{\circ} \text{ N}, 100-120^{\circ} \text{ E})$, the temperature gradient at 925 hPa was as high as 4.9 C per degree. Strong ascents occurred ahead of the front, with vertical velocity being ~ 0.05 m s⁻¹ at 900 hPa and ~ 0.2 m s⁻¹ at 750 hPa, and increasing with altitude until 300–250 hPa where the maximum vertical velocity was 0.259 m s⁻¹. Consequently, the high CO can be rapidly lifted to the upper troposphere in this case.

FLEXPART was also used to trace down high CO in the MOPITT image by releasing air particles in the boxed area in Fig. 6c (indicated by a bar in Fig. 4c). We found that the most CO came from the southwest part of China (boxed area in Fig. 13) where MO-PITT CO composite of 3–10 April 2005 shows high CO of 250–300 ppbv. This CO was



lifted along the WCB described above. This agrees with the GEOS-Chem simulation which shows the major contribution of the anthropogenic source to CO in the upper troposphere, likely from the Sichuan Basin (Fig. 15d).

5 Discussion

- In the three CO episodes, high CO abundances above 300 ppbv were observed in aircraft MOZAIC data in the free troposphere over East Asia during 2003–2005. In cases 2003 and 2004, the peak value reached 500 ppbv. In case 2005, CO concentration of 300 ppbv was observed at the upper troposphere (~ 300 hPa). These are among the highest CO abundances which have been documented at these altitudes in East Asia.
- Ding et al. (2009) observed high CO episode of ~ 1185 ppbv at 2.6 km (850–700 hPa) over the North China Plain in summer 2007. Nedelec et al. (2005) found CO up to 800 ppbv above 8 km (~ 400 hPa) near the fire region of Lake Baikal on 3 and 4 June 2003. Highest CO concentrations during TRACE-P were between 250–300 ppbv from 2–12 km (Heald et al., 2003; Liu et al., 2003; Miyazaki et al., 2003). Occurrences of such high CO episodes are not by chance. They reflect the uniqueness and complexity

of meteorology, orography, vegetation covers, and CO sources in East Asia. The possibility of occurrences of such high CO is further illustrated in Table 2. As the three cases ocurred near Japan, MOZAIC data around Narita and its surrounding areas from 2001 to 2006 are summarized, showing occurrences of various CO abundance

- ²⁰ ranges in the boundary layer (the surface-850 hPa), the lower (850–600 hPa), middle (600–400 hPa), and upper (400–200 hPa) troposphere. Among all the data in the upper troposphere, CO abundances occurred 93 times (17%) between 200–300 ppbv, 19 times (4%) between 300–400 ppbv, and 6 times (1%) over 400 ppbv. In the middle troposphere, the fraction of occurrences of CO within 200–300, 300–400, and over
- ²⁵ 400 ppbv was 14, 3, and 2%, respectively. In the boundary layer, the highest occurrences of CO abundances (38% of all the data in the layer) were within 200–300 ppbv, while the range was within 100–200 ppbv in the lower (47%), middle (74%), and upper



troposphere (66%). Seasonally, there were more high CO occurrences in the higher altitudes in spring and summer than in fall and winter.

This study illustrates that the topography in East Asia can affect vertical transport of CO in different ways. East Asia's topography varies significantly across its vast width,
⁵ increasing from east to west, with a variety of terrains. In addition to its general function in orograpic lifting (in case 2004), topography can also interplay with frontal systems and enhance the uplifting substantially in the North China Plain (in cases 2004 and 2005). Under the influence of the Tibetan Plateau, the southwest vortex (or the Sichuan low) is formed (Tao and Ding, 1981) and can facilitate strong convection in the Sichuan
¹⁰ basin (in case 2005). In particular, topography-induced convection due to the leeside troughs east of the Hengduan Mountains, first proposed by Lin et al. (2009), offers a new mechanism for vertical transport of pollutions from the region (in case 2004).

Extratropical cyclones and associated frontal activities are important in lifting CO from the boundary layer to the free troposphere. This can be harmful to regions downwind because the lifted CO can be transported for a long distance by strong upper level

- wind because the lifted CO can be transported for a long distance by strong upper level winds (Stohl et al., 2001, 2002; Damoah et al., 2004). This also applies to other air pollution. Zhao et al. (2008) found that the influence of Asian dust storms on North American ambient particulate matter levels is highly related to the height to which the frontal cyclones in East Asia can lift dust. This study shows that when air masses with high
- CO coincide with the strongest part of a WCB, uplifting of CO to the upper troposphere is possible. East Asia is one of two regions between 25–45° N with most frequent WCB events (Eckhardt et al., 2004). Inside East Asia, there are two regions where cyclones occur most frequently: one over the lee sides of the Altai-Sayan and the other in the East China Sea and the Sea of Japan, occurring mostly in spring and summer over
- ²⁵ both regions (Chen et al., 1991; Yue and Wang, 2008). The seasons and locations of three cases just match well with these two areas and active cyclone seasons, which should not be taken by chance. These are the locations and seasons where and when we can expect similar events to happen in the future. Chen et al. (1991) suggested a decline in cyclonic events in East Asia from 1957 to 1977 and no such decline from



1977 to 1987. Recently, an analysis for a longer term from 1951 to 2010 based on ensembles of Twentieth Century Reanalysis (20CR) showed a decreasing trend in the northern part of the Sea of Japan and an increasing trend over the southern part of the Sea of Japan and the leeside of the Altai-Sayan in summer (Wang et al., 2013). The implications of these trends on uplifting of CO deserve further studying.

Biomass burning is identified as an important source for all three episodes, suggesting that CO from sporadic fire activities can provide an additional source to the less varying anthropogenic emissions and enhance chances of high CO episodes. The fire regions shown in this study are the places with dense vegetation covers and with most active forest fires in East Asia (Schultz, 2002; Duncan et al., 2003). These fires are usually most active in summer in boreal forest in Russia, like in case 2003, and in spring in the southern East Asia, like cases 2004 and 2005.

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GEOS-Chem and FLEXPART simulations reveal different CO signatures from biomass burning and anthropogenic sources in the high CO episodes, reflecting dif-

- ¹⁵ ferent transport pathways and mechanisms and locations of both sources. In case 2003, CO from large forest fires near Lake Baikal dominated the elevated CO, which had been rapidly transported upward by a frontal system from the fire plumes. In case 2004, anthropogenic CO from the North China Plain experienced frontal lifting and mostly reached ~ 700 hPa near the East China Sea, while CO from biomass burning
- experienced orographic lifting, leeside-troughs induced convection, and frontal lifting through two separate transport pathways, leading to two distinct CO enhancements around 700 hPa and 300 hPa. In case 2005, along a WCB in the East China Sea and the Sea of Japan, anthropogenic CO from the North China Plain and from the Sichuan basin prevailed in the northern and southern part of the WCB, while CO from
- ²⁵ biomass burning in Indochina and southern China was mostly distributed in the middle. The source allocation is very sensitive to the location of the front. Overall, the anthropogenic sources contributed more than biomass burning to the CO in the middle and upper troposphere in this case. It would be helpful to conduct statistical analysis of the CO source distribution along WCBs in East Asia in the future.



MOPITT satellite data can further extend MOZAIC CO observations to larger areas and over wider altitude ranges. High CO concentrations were observed by MOPITT over thousand kilometers horizontally and a few kilometers vertically on the synoptic scale. Such variations of CO are usually diluted on longer time scales. Earlier applica-

tions of MOPITT CO data over East Asia were mostly focused on monthly or seasonal CO variations (Tanimoto et al., 2008; Zhao et al., 2010; Hao et al., 2011; Liu et al., 2011; Zhou et al., 2013; Su et al., 2014). The new understanding of synoptic disturbances from MOPITT and MOZAIC data in this study can help improve simulation of synoptic processes in atmospheric models, eventually leading to more realistic chemi cal weather forecast in the future (Lawrence et al., 2003)

MOPITT CO retrievals were validated with the MOZAIC aircraft observations at four cities in East Asia after taking the MOPITT averaging kernels and the a priori into consideration. In the upper troposphere, MOPITT V5 retrievals are usually higher than the MOZAIC measurements, while in the lower troposphere, MOPITT retrievals are higher than MOZAIC measurements in Beijing and Narita but lower at Shanghai and Hong Kong. The mean relative biases in all the MOPITT retrieval levels are within 20%, with the lowest bias around 500–400 hPa (close to zero). The DFS of MOPITT V5 is

generally enhanced compared with the V4 product.

While MOPITT provides unprecedented new datasets for a better understanding of the atmospheric chemistry, the vertical variation in CO seemed not to be fully resolved

- the atmospheric chemistry, the vertical variation in CO seemed not to be fully resolved in earlier applications of MOPITT data (Jacob et al., 2003). Nevertheless, some studies suggest that MOPITT indeed has some vertical sensitivity. Kar et al. (2004) found Asian summer monsoon plumes in MOPITT CO data as a strong enhancement of CO in the upper troposphere over India and southern China. Deeter et al. (2004) illustrated
- similar distributions of the rain rate and the ratio of MOPITT CO at 350 to at 850 hPa in the Tropical Eastern Pacific Ocean. Liu et al. (2006) observed large differences (20–40 ppbv) in MOPITT CO at 250 hPa between two cases of vertical transport of CO and attributed the differences to the respective weather systems. In this study, MOPITT's vertical sensitivity with the new V5 retrievals is well illustrated in three cases with high



CO episodes (Figs. 5 and 6). In Fig. 5, the smoothed MOZAIC profiles were calculated using the averaging kernels and the a priori in an area upwind of the MOZAIC measurement within $0-5^{\circ}$ distance for each case as there were no MOPITT data available at the locations of the MOZAIC measurements. Although this may introduce some bias,

- the smooth MOZAIC profiles in V5 show more vertical structure in CO than an earlier version of MOPITT data in Jacob et al. (2003). Because the CO retrieval at a certain pressure level is often contaminated by CO from other levels, in other words, the averaging kernel matrix is less than I, MOPITT cannot specify the exact height of elevated CO shown in the MOZAIC measurement. The magnitude of elevated CO in MOPITT
- retrievals was much lower than in the MOZAIC data at altitudes where CO peaked and thus the vertical variation of CO was still much smoothed in MOPITT data. However, spatial variation in MOPITT CO data can show elevated CO in the lower, middle, and upper troposphere in the respective cases. The strong CO source in case 2003 is reflected differently from the other two cases. The vertical distribution of CO over large
- ¹⁵ area and in a wide range of altitudes shown in Fig. 6 for the three cases matches with simulations of GEOS-Chem and FLEXPART, sometimes remarkably well. The detection of high CO in the upper troposphere in case 2005 makes MOPITT data promising in studying vertical transport of CO or the vertical distribution of CO qualitatively up to that level. One limitation for MOPITT's application of vertical transport is the com-
- plication of clouds, which often accompany frontal systems. As shown in cases 2004 and 2005, CO is usually high in cloudy areas. Therefore, the magnitude of CO abundances can be statistically underestimated by MOPITT in these areas. Large gaps due to clouds and the limited MOPITT swath make it challenging to use MOPITT data for studying synoptic processes.
- Pollution transport can be tracked computationally with Euler and Lagrangian approaches, as represented by GEOS-Chem and FLEXPART models, respectively. The two models have successfully reproduced the observed aircraft and satellite CO to some extent. GEOS-Chem can not only track transport of CO (a physical process) but also consider chemical reactions during the transport while FLEXPART can visualize



transport pathways and pin down source regions effectively, without considering chemical functions in the meantime. GEOS-Chem can also fill the gaps in MOPITT satellite data (Figs. 10, 11, 14). GEOS-Chem simulates cases 2004 and 2005 well but cannot reproduce the magnitude of the elevated CO in MOZAIC data in case 2003, possibly
 ⁵ because its parameterization of large forest fires was conservative or the fire emissions were underestimated in the inventory. Nassar et al. (2009) reported underestimates of CO by GEOS-Chem over the 2006 Indonesia fire region, in comparison with the Tropospheric Emission Spectrometer (TES) observations. FLEXPART can generally simulate the three cases, strikingly well sometimes in agreement with observed details in space and time, although discrepancies between ELEXPART and satellite and aircraft

¹⁰ space and time, although discrepancies between FLEXPART and satellite and aircraft observations can be found in various places on small scales.

6 Conclusions

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East Asia is characterized by its unique and complex meteorology, topography, vegetation covers, and CO sources. The characteristics are reflected in uplifting of CO illustrated in three high CO episodes during 2003–2005 in this study. Through integrated analyses of observations from the MOZAIC aircraft and the MOPITT satellite and simulations from a trajectory dispersion model FLEXPART (Stohl et al., 2005) and a chemical transport model GEOS-Chem (Bey et al., 2001), this study draws the major conclusions as follows.

- In the three CO episodes, high CO abundances of 300–550 ppbv are observed in aircraft MOZAIC data in the free troposphere. These are among the highest CO abundances ever documented at these altitudes. The three cases occurred in the seasons and at locations where meteorological and CO source conditions are favorable for such episodes.
- 25 2. GEOS-Chem and FLEXPART simulations reveal different CO signatures from biomass burning and anthropogenic sources in the CO enhancement in the three



cases, reflecting different transport pathways and mechanisms and locations of both sources. In case 2003, CO from large forest fires near Lake Baikal dominated the elevated CO. In case 2004, anthropogenic CO came from the North China Plain and mostly reached ~ 700 hPa near the East China Sea, while CO from biomass burning from Indochina was transported through two separate pathways, leading to two distinct CO enhancements around 700 hPa and 300 hPa. In case 2005, along a WCB over the East China Sea and the Sea of Japan, anthropogenic CO from the North China Plain and from the Sichuan basin prevailed in the northern and southern part of the WCB, while CO from biomass burning in Indochina and southern China was mostly distributed in the middle part of the WCB.

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3. Topography in East Asia influences vertical transport of CO in different ways. In particular, topography-induced leeside troughs east of the Hengduan Mountains over Indochina lead to strong convection. This new mechanism proposed by Lin et al. (2009) is confirmed and further extended in this study. Strong convection from the Sichuan basin also plays an important role in transporting anthropogenic CO to the free troposphere. The topography interplaying with frontal activates can enhance the vertical transport of CO substantially in North China Plain.

4. Extratropical cyclones and associated frontal activities are important mechanism in lifting CO from the boundary layer to the free troposphere, as illustrated by the three cases and many earlier studies. East Asia is one of two regions between 25– 45° N with most frequent WCB events (Eckhardt et al., 2004). Inside East Asia, there are two regions where cyclones occur most frequently: one over the lee sides of the Altai-Sayan and the other in the East China Sea and the Sea of Japan, occurring mostly in spring and summer over both regions (Chen et al., 1991). The seasons and locations of the three high CO episodes just match well with these two areas and active cyclone seasons, which may not happen by chance.



5. Biomass burning is identified as an important source for all three episodes, suggesting that CO from sporadic fire activities can provide additional CO to less varying anthropogenic emission and enhance chances of high CO episodes. The fire regions shown in this study are the places with dense vegetation covers and with most active forest fires in East Asia.

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- 6. The MOPITT's vertical sensitivity is enhanced in its new V5 NIR/TIR data. The daytime V5 data can detect synoptic disturbance of weather systems on horizon-tal variation of CO. The data also show more vertical structure than earlier versions and can distinguish CO enhancements at different layers of the troposphere, although the detected high CO is over a broad range in altitudes and lacks detailed vertical structure in comparison with the aircraft observations. Because the CO retrieval at a certain pressure level is often contaminated by CO from other levels, the MOPITT retrievals usually underestimate elevated CO peaks during the high CO episodes. The complication of clouds within frontal systems can generate large gaps in MOPITT data and cause underestimation of CO statistically in these regions. Nevertheless, MOPITT data may be used to qualitatively help diagnose vertical transport processes, with caution on their absolute CO values. In general, MOPITT substantially underestimates CO in high CO episodes and, on average, slightly overestimats the background CO in the upper troposphere.
- Acknowledgements. The authors gratefully acknowledge the following data and modeling tools. The satellite CO data are provided by the MOPITT team and acquired from the NASA Langley Research Center Atmospheric Science Data Center. The MOZAIC CO data are from the European Commission, Airbus, and the Airlines (Lufthansa, Austrian, Air France) who carry free of charge the MOZAIC equipment and perform the maintenance since 1994. The Final Analnic Dete (SNI).
- ysis Data (FNL) were obtained from NOAA CDC. The GEOE-Chem model is developed and managed by the Atmospheric Chemistry Modeling Group at Harvard University with support from the NASA Atmospheric Chemistry Modeling and Analysis Program (ACMAP). The FLEX-PART model development team consists of Andreas Stohl, Sabine Eckhardt, Harald Sodemann, and John Burkhart at the Norwegian Institute for Air Research (NILU). Financial support is provided by an energy fund from the Institute of Remete Sensing and Digital Forth Chinage
- ³⁰ is provided by an open fund from the Institute of Remote Sensing and Digital Earth, Chinese



Academy of Sciences (OFSLRSS201107), the Key Basic Research Program (2010CB950704, 2014CB441203), and the Natural Science Foundation of China (41375140).

References

5

Banic, C. M., Isaac, G. A., Cho, H. R., and Iribane, J. V.: The distribution of pollutants near a frontal surface: a comparison between field experiment and modeling, Water Air Soil Poll.,

- a frontal surface: a comparison between field experiment and modeling, water Air Soil Poll., 30, 171–177, 1986.
- Barret, B., Le Flochmoen, E., Sauvage, B., Pavelin, E., Matricardi, M., and Cammas, J. P.: The detection of post-monsoon tropospheric ozone variability over south Asia using IASI data, Atmos. Chem. Phys., 11, 9533–9548, doi:10.5194/acp-11-9533-2011, 2011.
- Berntsen, T. K., Karlsdóttir, S., and Jaffe, D. A.: Influence of Asian emissions on the composition of air reaching the north western United States, Geophys. Res. Lett., 26, 2171-2174, doi:10.1029/1999GL900477, 1999.
 - Bertschi, I. B., Jaffe, D. A., Jaeglé, L., Price, H. U., and Dennison, J. B.: PHOBEA/ITCT 2002 airborne observations of trans-Pacific transport of ozone, CO, VOCs and aerosols to the
- northeast Pacific: impacts of Asian anthropogenic and Siberian Boreal fire emissions, J. Geophys. Res., 109, D23S12,doi:10.1029/2003JD004328, 2004.
 - Bethan, S., Vaughan, G., Gerbig, C., Volz-Thoms, A., Richer, H., and Tiddeman, D. A.: Chemical air mass differences near fronts, J. Geophys. Res., 103, 13413–13434, 1998.

Bey, I., Jacob, D. J., Yantosca, R. M., Logan, J. A., Field, B. D., Fiore, A. M., Li, Q., Liu, H. Y.,

- Mickley, L. J., and Schultz, M. G.: Global modeling of tropospheric chemistry with assimilated meteorology: model description and evaluation, J. Geophys. Res., 106, 23073–23095, 2001. Brown, R. M., Daum, P. H., Schwartz, S. E., and Hjelmfelt, M. R.: Variations in the chemical composition of clouds during frontal passage, in: The Meteorology of Acid Deposition, edited by: Samson, P. J., Air Pollut. Control Assoc., Pittsburgh, Pa., 202–212, 1984.
- ²⁵ Chan, D., Yuen, C. W., Higuchi, K., Shashkov, A., Liu, J., Chen, J., and Worthy, D.: On the CO₂ exchange between the atmosphere and the biosphere: the role of synoptic and mesoscale processes, Tellus B, 56, 194–212, 2004.
 - Chen, B., Xu, X. D., Yang, S., and Zhao, T. L.: Climatological perspectives of air transport from atmospheric boundary layer to tropopause layer over Asian monsoon regions during



28051

boreal summer inferred from Lagrangian approach, Atmos. Chem. Phys., 12, 5827–5839, doi:10.5194/acp-12-5827-2012, 2012.

- Chen, S., Kuo, Y., Zhong, P., and Bai, Q.: Synoptic climatology of cyclogenesis over East Asia, 1958–1987, Mon. Weather Rev., 119, 1407–1418, 1991.
- ⁵ Chung, K. K., Chan, J. C. L., Ng, C. N., Lam, K. S., and Wang, T.: Synoptic conditions associated with high carbon monoxide episodes at coastal station in Hong Kong, Atmos. Environ., 33, 3099–3095, 1999.

Cooper, O. R., Moody, J. L., Parrish, D. D., Trainer, M., Ryerson, T. B., Holloway, J. S., Hübler, G., Fehsenfeld, F. C., and Evans, M. J.: Trace gas composition of midlatitude cy-

clones over the western North Atlantic Ocean: a conceptual model, J. Geophys. Res., 107, D7, doi:10.1029/2001JD000901, 2002.

Cooper, O. R., Forster, C., Parrish, D., Dunlea, E., Habler, G., Fehsenfeld, F., Holloway, J., Oltmans, S., Johnson, B., Wimmers, A., and Horowitz, L.: On the life-cycle of a stratospheric intrusion and its dispersion into polluted warm conveyor belts, J. Geophys. Res., 109, D23S09, doi:10.1029/2003JD004006. 2004.

Cooper, O. R., Stohl, A., Hubler, G., Hsie, E. Y., Parrish, D. D., Tuck, A. F., Kiladis, G. N., Oltmans, S. J., Johnson, B. J., Shapiro, M., Moody, J. L., and Lefohn, A. S.: Direct transport of midlatitude stratospheric ozone into the lower troposphere and marine boundary layer of the tropical Pacific Ocean, J. Geophys. Res., 110, D23310, doi:10.1029/2005JD005783, 2005.

15

- Cooper, O. R., Stohl, A., Trainer, M., Thompson, A., Witte, J. C., Oltmans, S. J., Johnson, B. J., Merrill, J., Moody, J. L., Tarasick, D., N'ed'elec, P., Forbes, G., Newchurch, M. J., Schmidlin, F. J., Johnson, B. J., Turquety, S., Baughcum, S. L., Ren, X., Fehsenfeld, F. C., Meagher, J. F., Spichtinger, N., Brown, C. C., McKeen, S. A., McDermid, I. S., and Leblanc, T.:
- Large upper tropospheric ozone enhancements above mid-latitude North America during summer: in situ evidence from the IONS and MOZAIC ozone monitoring network, J. Geophys. Res., 111, D24S05, doi:10.1029/2006JD007306, 2006.

Cristofanelli, P., Bonasoni, P., Collins, W., Feichter, J., Forster, C., James, P., Kentarchos, A., Kubik, P. W., Land, C., Meloen, J., Roelofs, G. J., Siegmund, P., Sprenger, M., Schnabel, C.,

³⁰ Stohl, A., Tobler, L., Tositti, L., Trickl, T., and Zanis, P.: Stratosphere-to-troposphere transport: a model and method evaluation, STACCATO special section of J. Geophys. Res., 108, 8525, doi:10.1029/2002JD002600, 2003.

Daley, R.: Atmospheric Data Analysis, Cambridge University Press, Cambridge, 1991.



- Damoah, R., Spichtinger, N., Forster, C., James, P., Mattis, I., Wandinger, U., Beirle, S., Wagner, T., and Stohl, A.: Around the world in 17 days - hemispheric-scale transport of forest fire smoke from Russia in May 2003, Atmos. Chem. Phys., 4, 1311–1321, doi:10.5194/acp-4-1311-2004, 2004.
- ⁵ Davies, D. K., Ilavajhala, S., Wong, M. M., and Justice, C. O.: Fire information for resource management system: archiving and distributing MODIS active fire data, IEEE T. Geosci. Remote, 47, 72–79, 2009.

Deeter, M. N., Emmons, L. K., Francis, G. L., Edwards, D. P., Gille, J. C., Warner, J. X., Khattatov, B., Ziskin, D., Lamarque, J.-F., Ho, S.-P., Yudin, V., Attié, J.-L., Packman, D.,

¹⁰ Chen, J., Mao, D., and Drummond, J. R.: Operational carbon monoxide retrieval algorithm and selected results for the MOPITT instrument, J. Geophys. Res., 108, 4399, doi:10.1029/2002JD003186, 2003.

Deeter, M. N., Emmons, L. K., Edwards, D. P., Gille, J. C., and Drummond, J. R.: Vertical resolution and information content of CO profiles retrieved by MOPITT, Geophys. Res. Lett., 21 J 15110, doi:10.1000/000401.000005.0004

¹⁵ 31, L15112, doi:10.1029/2004GL020235, 2004.

Deeter, M. N., Worden, H. M., Edwards, D. P., Gille, J. C., and Andrews, A. E.: Evaluation of MO-PITT retrievals of lower-tropospheric carbon monoxide over the United States, J. Geophys. Res., 117, D13306, doi:10.1029/2012JD017553, 2012.

Dickerson, R. R., Huffman, G. J., Luke, W. T., Nunnermacker, L. J., Pickering, K. E.,

Leslie, A. C. D., Lindsey, C. G., Slinn, W. G. N., Kelly, T. J., Daum, P. H., Delany, A. C., Greenberg, J. P., Zimmerman, P. R., Boatman, J. F., Ray, J. D., and Stedman, D. H.: Thunderstorms – an important mechanism in the transport of air pollutants, Science, 235, 4787, 460–464, 1987.

Dickerson, R. R., Li, C., Li, Z., Marufu, L., T., Stehr, J. W., McClure, B., Krotkov, N., Chen, H.,

- ²⁵ Wang, P., Xia, X., Ban, X., Gong, F., Yuan, J., and Yang, J.: Aircraft observations of dust and pollutants over northeast China: insight into the meteorological mechanisms of transport, J. Geophys. Res., 112, D24S90, doi:10.1029/2007JD008999, 2007.
 - Ding, A., Wang, T., Xue, L., Gao, J., Stohl, A., Lei, H., Jin, D., Ren, Y., Wang, X., Wei, X., Qi, Y., Liu, J., and Zhang, X.: Transport of north China air pollution by midlatitude cyclones:
- case study of aircraft measurements in summer 2007, J. Geophys. Res., 114, D08304, doi:10.1029/2008JD011023, 2009.



- Donnell, E. A., Fish, D. J., Dicks, E. M., and Thorpe, A. J.: Mechanisms for pollutant transport between the boundary layer and the free troposphere, J. Geophys. Res., 106, 7847–7856, 2001.
- Drummond, J. R.: Measurements of pollution in the troposphere (MOPITT), in: The Use of EOS
- ⁵ for Studies of Atmospheric Physics, edited by: Gille, J. C. and Visconti, G., North Holland, New York, 77–101, 1992.
 - Drummond, J. R. and Mand, G. S.: The measurements of pollution in the troposphere (MOPITT) instrument: overall performance and calibration requirements, J. Atmos. Ocean. Tech., 13, 314–320, 1996.
- ¹⁰ Duncan, B. N., Martin, R. V., Staudt, A. C., Yevich, R., and Logan, J. A.: Interannual and seasonal variability of biomass burning emissions constrained by satellite observations, J. Geophys. Res., 108, 4040, doi:10.1029/2002JD002378, 2003.
 - Duncan, B. N., Logan, J. A., Bey, I., Megretskaia, I. A., Yantosca, R. M., Novelli, P. C., Jones, N. B., and Rinsland, C. P.: Global budget of CO, 1988–1997: source estimates and
- validation with a global model, J. Geophys. Res., 112, D22301, doi:10.1029/2007JD008459, 2007.
 - Eckhardt, S., Stohl, A., Wernli, H., James, P., Forster, C., and Spichtinger, N.: A 15-Year climatology of warm conveyor belts, J. Climate, 17, 218–237, 2004.

Edwards, D. P., Halvorson, C. M., and Gille, J. C.: Radiative transfer modeling for the EOS Terra satellite measurements of pollution in the troposphere (MOPITT instrument), J. Geophys.

Res., 104, 16755–16775, 1999.

20

- Emmons, L. K., Deeter, M. N., Gille, J. C., Edwards, D. P., Attie, J.-L., Warner, J., Ziskin, D., Khattatov, B., Yudin, V., Lamarque, J.-F., Ho, S.-P., Mao, D., Chen, J. S., Drummond, J., Novelli, P., Sachse, G., Coffey, M. T., Hannigan, J. W., Gerbig, C., Kawakami, S., Kondo, Y.,
- ²⁵ Takegawa, N., Baehr, J., and Ziereis, H.: Validation of MOPITT CO retrievals with aircraft in situ profiles, J. Geophys. Res., 109, D03309, doi:10.1029/2003JD004101, 2004.
 - Giglio, L., Descloitres, J., Justice, C. O., and Kaufman, Y. J.: An enhanced contextual fire detection algorithm for MODIS, Remote Sens. Environ., 87, 273–282, 2003.
- Hao, H., Valks, P., Loyola, D., Chen, Y. F., and Zimmer, W.: Space-based measure ments of air quality during the World Expo 2010 in Shanghai, Environ. Res. Lett., 6, 044004,doi:10.1088/1748-9326/6/4/044004, 2011.
 - He, H., Tarasick, D. W., Hocking, W. K., Carey-Smith, T. K., Rochon, Y., Zhang, J., Makar, P. A., Osman, M., Brook, J., Moran, M. D., Jones, D. B. A., Mihele, C., Wei, J. C., Osterman, G.,



Argall, P. S., McConnell, J., and Bourqui, M. S.: Transport analysis of ozone enhancement in Southern Ontario during BAQS-Met, Atmos. Chem. Phys., 11, 2569–2583, doi:10.5194/acp-11-2569-2011, 2011.

Heald, C. L., Jacob, D. J., Fiore, A. M., Emmons, L. K., Gille, J. C., Deeter, M. N., Warner, J.,

- Edwards, D. P., Crawford, J. H., Hamlin, A. J., Sachse, G. W., Browell, E. V., Avery, M. A., Vay, S. A., Westberg, D. J., Blake, D. R., Singh, H. B., Sandholm, S. T., Talbot, R. W., and Fuelberg, H. E.: Asian outflow and transpacific transport of carbon monoxide and ozone pollution: an integrated satellite, aircraft and model perspective, J. Geophys. Res., 108, 4804, doi:10.1029/2003JD003507, 2003.
- Hocking, W. K., Carey-Smith, T. K., Tarasick, D. W., Argall, P. S., Strong, K., Rochon, Y., Zawadzki, I., and Taylor, P. A.: Detection of stratospheric ozone intrusion by wind profiler radars, Nature, 450, 281–284, doi:10.1038/nature06312, 2007.
 - Holloway, T., Levy II, H., and Kasibhatla, P.: Global distribution of carbon monoxide, J. Geophys. Res., 105, 12123–12147, doi:10.1029/1999JD901173, 2000.
- ¹⁵ Jacob, D. J.: Introduction to Atmospheric Chemistry, Princeton University Press, Princeton, New Jersey, 1999.
 - Jacob, D. J., Crawford, J. H., Kleb, M. M., Connors, V. S., Bendura, R. J., Raper, J. L., Sachse, G. W., Gille, J. C., Emmons L., and Heald, C. L.: Transport and Chemical Evolution over the Pacific (TRACE-P) aircraft mission: design, execution, and first results, J. Geophys. Res., 108, 9000, doi:10.1029/2002JD003276, 2003.
 - Jaffe, D., Anderson, T., Covert, D., Kotchenruther, R., Trost, B., Danielson, J., Simpson, W., Berntsen, T., Karlsdottir, S., Blake, D., Harris, J., Carmichael, G., and Uno, I.: Transport of Asian air pollution to North America, Geophys. Res. Lett., 26, 711–714, 1999.

20

Jaffe, D., Bertschi, I., Jaegle, L., Novelli, P., Reid, J. S., Tanimoto, H., Vingarzan, R.,and

- Westphal, D. L.: Long-range transport of Siberian biomass burning emissions and impact on surface ozone in western North America, Geophys. Res. Lett., 31, L16106, doi:10.1029/2004GL020093, 2004.
 - Jiang, Z., Jones, D. B. A., Kopacz, M., Liu, J., Henze, D. K., and Heald, C.: Quantifying the impact of model errors on top-down estimates of carbon monoxide emissions using satellite
- ³⁰ observations, J. Geophys. Res. 116, D15306, doi:10.1029/2010JD015282, 2011.
 - Jones, D. B. A., Bowman, K. W., Logan, J. A., Heald, C. L., Liu, J., Luo, M., Worden, J., and Drummond, J.: The zonal structure of tropical O₃ and CO as observed by the Tropospheric



28055

for field campaign planning: predictions and observations of large-scale features during MI-

Res., 105, 26871-26890, doi:10.1029/2000JD900180, 2000. Lawrence, M. G., Rasch, P. J., von Kuhlmann, R., Williams, J., Fischer, H., de Reus, M., Lelieveld, J., Crutzen, P. J., Schultz, M., Stier, P., Huntrieser, H., Heland, J., Stohl, A.,

30 Forster, C., Elbern, H., Jakobs, H., and Dickerson, R. R.: Global chemical weather forecasts

from the boundary layer to the free atmosphere, Geophys. Res. Lett., 28, 2907–2910, 2001. 25 Lavoué, D., Liousse, C., Cachier, H., Stocks, B. J., and Goldammer, J. G.: Modeling of carbonaceous particles emitted by boreal and temperate wildfires at northern latitudes, J. Geophys.

timates of CO sources with high resolution by adjoint inversion of multiple satellite datasets (MOPITT, AIRS, SCIAMACHY, TES), Atmos. Chem. Phys., 10, 855-876, doi:10.5194/acp-10-855-2010, 2010. Kowol-Santen, J., Beekmann, M., Schmitgen, S., and Dewey, K.: Tracer analysis of transport

Kopacz, M., Jacob, D. J., Fisher, J. A., Logan, J. A., Zhang, L., Megretskaia, I. A., Yantosca, R. M., Singh, K., Henze, D. K., Burrows, J. P., Buchwitz, M., Khlystova, I., McMillan, W. W., Gille, J. C., Edwards, D. P., Eldering, A., Thouret, V., and Nedelec, P.: Global es-

mour, M. D., Edwards, D. P., Deeter, M. N., Gille, J. C., and Richter, A.: Measurement of low-altitude CO over the Indian subcontinent by MOPITT, J. Geophys. Res., 113, D16307,

wards, D. P., Deeter, M. N.: Carbon monoxide (CO) maximum over the Zagros mountains in the Middle East: signature of mountain venting?, Geophys. Res. Lett., 33, L15819, doi:10.1029/2006GL026231.2006. Kar, J., Jones, D. B. A., Drummond, J. R., Attie, J. L., Liu, J., Zou, J., Nichitiu, F., Sey-

15

20

doi:10.1029/2007JD009362, 2008.

- Gille, J. C., Edwards, D. P., Deeter, M. N., Francis, G., Ziskin, D., and Warner, J.: Evidence of vertical transport of carbon monoxide from measurements of pollution in the troposphere (MOPITT). Geophys. Res. Lett., 31, L23105, doi:10.1029/2004GL021128, 2004. 10 Kar, J., Drummond, J. R., Jones, D. B. A., Liu, J., Nichitiu, F., Zou, J., Gille, J. C. Ed-
- 83, 244–262, 2002. 5 Kar, J., Bremer, H., Drummond, J. R., Rochon, Y. J., Jones, D. B. A., Nichitiu, F., Zou, J., Liu, J.,

Atmos. Chem. Phys., 9, 3547–3562, doi:10.5194/acp-9-3547-2009, 2009. Justice, C. O., Giglio, L., Korontzi, S., Owens, J., Morisette, J. T., Roy, D., Descloitres, J., Alleaume, S., Petitcolin, F., and Kaufman, Y.: The MODIS fire products, Remote Sens. Environ.,

Emission Spectrometer in November 2004 – Part 1: Inverse modeling of CO emissions,

Discussion

Paper





14, 28019–28077, 2014

Discussion Paper Uplifting of carbon monoxide to the free troposphere in East Discussion Asia K. Ding et al. Paper **Title Page** Abstract **Discussion** Paper Conclusions Tables **Figures**

NOS, CONTRACE, and INDOEX, Atmos. Chem. Phys., 3, 267–289, doi:10.5194/acp-3-267-2003, 2003.

- Li, Q. B., Jacob, D. J., Park, R. J., Wang, Y. X., Heald, C. L., Hudman, R., Yantosca, R. M., Martin, R. V., and Evans, M. J.: North American pollution outflow and the trapping of convectively lifted pollution by upper-level anticyclone, J. Geophys. Res., 110, D10301, doi:10.1029/2004JD005039, 2005.
 - Li, Z., Chen, H., Cribb, M., Dickerson, R., Holben, B., Li, C., Lu, D., Luo, Y., Maring, H., Shi, G., Tsay, S.-C., Wang, P., Wang, Y., Xia, X., Zheng, Y., Yuan, T., and Zhao, F.: Preface to special section on East Asian Studies of Tropospheric Aerosols: an International Regional Experiment (EAST-AIRE), J. Geophys. Res., 112, D22S00, doi:10.1029/2007JD008853, 2007.
- ment (EAST-AIRE), J. Geophys. Res., 112, D22S00, doi:10.1029/2007JD008853, 2007. Liang, Q., Jaegle, L., Jaffe, D. A., Weiss-Penzias, P., Heckman, A., and Snow, J. A.: Longrange transport of Asian pollution to the northeast Pacific: seasonal variations and transport pathways of carbon monoxide, J. Geophys. Res., 109, D23S07, doi:10.1029/2003JD004402, 2004.
- Lin, C.-Y., Hsu, H.-m., Lee, Y. H., Kuo, C. H., Sheng, Y.-F., and Chu, D. A.: A new transport mechanism of biomass burning from Indochina as identified by modeling studies, Atmos. Chem. Phys., 9, 7901–7911, doi:10.5194/acp-9-7901-2009, 2009.
 - Liu, C., Beirle, S., Butler, T., Liu, J., Hoor, P., Jöckel, P., Penning de Vries, M., Pozzer, A., Frankenberg, C., Lawrence, M. G., Lelieveld, J., Platt, U., and Wagner, T.: Application
- of SCIAMACHY and MOPITT CO total column measurements to evaluate model results over biomass burning regions and Eastern China, Atmos. Chem. Phys., 11, 6083–6114, doi:10.5194/acp-11-6083-2011, 2011.
 - Liu, H. Y., Jacob, D. J., Bey, I., Yantosca, R. M., Duncan, B. N., and Sachse, G.W.: Transport pathways for Asian combustion outflow over the Pacific: interannual and seasonal variations, J. Geophys. Res., 108, 8786, doi:10.1029/2002JD003102, 2003.
 - Liu, J., Drummond, J. R., Li, Q., Gille, J. C., and Ziskin, D. C.: Satellite mapping of CO emission from forest fires in northwest America using MOPITT measurements, Remote Sens. Environ., 95, 502–516, 2005.

25

Liu, J., Drummond, J. R., Jones, D. B. A., Cao, Z., Bremer, H., Kar, J., Zou, J., Nichitiu, F., and

³⁰ Gille, J. C.: Large horizontal gradients in atmospheric CO at the synoptic scale as seen by spaceborne measurements of pollution in the troposphere, J. Geophys. Res., 111, D02306, doi:10.1029/2005JD006076, 2006.



Mari, C., Evans, M. J., Palmer, P. I., Jacob, D. J., and Sachse, G. W.: Export of Asian pollution during two cold front episodes of the TRACE-P experiment, J. Geophys. Res., 109, D15S17, doi:10.1029/2003JD004307, 2004.

Miyazaki, Y., Kondo, Y., Koike, M., Fuelberg, H. E., Kiley, C. M., Kita, K., Takegawa, N.,

Sachse, G. W., Flocke, F., Weinheimer, A. J., Singh, H. B., Eisele, F. L., Zondlo, M., Talbot, R. W., Sandholm, S. T., Avery, M. A., and Blake, D. R.: Synoptic-scale transport of reactive nitrogen over the western Pacific in spring, J. Geophys. Res., 108, 8788, doi:10.1029/2002JD003248,2003.

Marenco, A., Thouret, V., Nedelec, P., Smit, H., Helten, M., Kley, D., Karcher, F., Simon, P.,

- ¹⁰ Law, K., Pyle, J., Poschmann, G., Wrede, R. V., Hume, C., and Cook, T: Measurement of ozone and water vapor by Airbus in-service aircraft: the MOZAIC airborne program, An overview, J. Geophys. Res., 103, 25631–25642, 1998.
 - Nassar, R., Logan, J. A., Megretskaia, I. A., Murray, L. T., Zhang, L., and Jones, D. B. A.: Analysis of tropical tropospheric ozone, carbon monoxide, and water vapor during the 2006 El
- ¹⁵ Niño using TES observations and the GEOS-Chem model, J. Geophys. Res., 114, D17304, doi:10.1029/2009JD011760, 2009.
 - Nassar, R., Jones, D. B. A., Suntharalingam, P., Chen, J. M., Andres, R. J., Wecht, K. J., Yantosca, R. M., Kulawik, S. S., Bowman, K. W., Worden, J. R., Machida, T., and Matsueda, H.: Modeling global atmospheric CO₂ with improved emission inventories and CO₂
- production from the oxidation of other carbon species, Geosci. Model Dev., 3, 689–716, doi:10.5194/gmd-3-689-2010, 2010.
 - Nedelec, P., Thpuret, V., Brioude, J., Sauvage, B., Cammas, J., Stohl, A.: Extreme CO concentrations in the upper troposphere over northeast Asia in June 2003 from the in situ MOZAIC aircraft data, Geophys. Res. Lett., 32, L14807, doi:10.1029/2005GL023141, 2005.
- Novelli, P., Masarie, K. A., and Lang, P. M.: Distributions and recent changes of carbon monoxide in the lower troposphere, J. Geophys. Res., 103, 19015–19033, 1998.
 - Olivier, J. G. J. and Berdowski, J. J. M.: Global emission sources and sinks, in: The Climate System, edited by: Berdowski, J., Guicherit, R., and Heij, B. J., Swets & Zeitlinger, Lisse, the Netherlands, 33–77, 2001.
- Oltmans, S. J., Lefohn, A. S., Harris, J. M., Tarasick, D. W., Thompson, A. M., Wernli, H., Johnson, B. J., Novelli, P. C., Montzka, S. A., Ray, J. D., Patrick, L. C., Sweeney, C., Jefferson, A., Dann, T., Davies, J., Shapiro, M., Holben, B. N.: Enhanced ozone over western



28058

North America from biomass burning in Eurasia during April 2008 as seen in surface and profile observations, Atmos. Environ., 44, 4497-4509, 2010.

- Pan, L., Gille, J. C., Edwards, D. P., Bailey, P. L., and Rodgers, C. D.: Retrieval of tropospheric carbon monoxide for the MOPITT experiment, J. Geophys. Res., 103, 32277–32290, 1998.
- ⁵ Pickering, K. E., Dickerson, R. R., Huffman, G. J., Boatman, J. F., and Schanot, A.: Trace gas transport in the vicinity of frontal convective clouds, J. Geophys. Res., 93, 759-773, doi:10.1029/JD093iD01p00759, 1998.
 - Randel, W. J., Park, M., Emmons, L., Kinnison, D., Bernath, P., Walker, K. A., Boone, C., and Pumphrey, H.: Asian monsoon transport of pollution to the stratosphere, Science, 328, 611-

613. doi:10.1126/science.1182274. 2010. 10

Rogers, C. D.: Inverse Methods for Atmospheric Sounding, Theory and Practice, World Sci., River Edge, N.J., 2000.

Schultz, M. G.: On the use of ATSR fire count data to estimate the seasonal and interannual variability of vegetation fire emissions, Atmos. Chem. Phys., 2, 387-395, doi:10.5194/acp-2-387-2002, 2002,

15

20

- Stohl, A.: A 1-year Lagrangian "climatology" of airstreams in the North Hemisphere troposphere and lowermost stratosphere, J. Geophys. Res., 106, 7263-7279, 2001.
 - Stohl, A., Hittenberger, M., and Wotawa, G.: Validation of the Lagrangian particle dispersion model FLEXPART against large scale tracer experiment data, Atmos. Environ., 24, 4245-4264, 1998.
- Stohl, A., Eckhardt, S., Forster, C., James, P., and Spichtinger, N.: On the pathways and timescales of intercontinental air pollution transport, J. Geophys. Res., 107, 4684, doi:10.1029/2001JD001396, 2002.

Stohl, A., Forster, C., Frank, A., Seibert, P., and Wotawa, G.: Technical note: The Lagrangian

- particle dispersion model FLEXPART version 6.2, Atmos. Chem. Phys., 5, 2461-2474, 25 doi:10.5194/acp-5-2461-2005, 2005.
 - Streets, D. G., Zhang, Q., Wang, L., He, K., Hao, J., Wu, Y., Tang, Y., and Carmichael, G. R.: Revisiting China's CO emissions after the Transport and Chemical Evolution over the Pacific (TRACE-P) mission: synthesis of inventories, atmospheric modeling, and observations, J.
- Geophys. Res., 111, D14306, doi:10.1029/2006JD007118, 2006. 30
 - Su, M., Lin, Y., Fan, X., Peng, L., Zhao, C.: Impacts of global emissions of CO, NO_v, and CH₄ on China tropospheric hydroxyl free radicals, Adv. Atmos. Sci., 29, 4, 838-854, 2012.



- Suntharalingam, P., Jacob, D. J., Palmer, P. I., Logan, J. A., Yantosca, R. M., Xiao, Y., Evans, M. J., Streets, D., Vay, S. A., and Sachse, G.: Improved quantification of Chinese carbon fluxes using CO₂/CO correlations in Asian outflow, J. Geophys. Res., 109, D18S18, doi:10.1029/2003JD004362, 2004.
- ⁵ Tao, S. and Ding, Y.: Observational evidence of the influence of the Qinghai-Xizang (Tibet) Plateau on the occurrence of heavy rain and severe convective storms in China, B. Am. Meteorol. Soc., 62, 23–30, 1981.

Tanimoto, H., Sawa, Y., Yonemura, S., Yumimoto, K., Matsueda, H., Uno, I., Hayasaka, T., Mukai, H., Tohjima, Y., Tsuboi, K., and Zhang, L.: Diagnosing recent CO emissions and

¹⁰ ozone evolution in East Asia using coordinated surface observations, adjoint inverse modeling, and MOPITT satellite data, Atmos. Chem. Phys., 8, 3867–3880, doi:10.5194/acp-8-3867-2008, 2008.

Tsutsumi, Y., Makino, Y., and Jensen, J. B.: Vertical and latitudinal distributions of tropospheric ozone over the western Pacific: case studies from the PACE aircraft missions, J. Geophys. Res., 108, 4251, doi:10.1029/2001JD001374, 2003.

- ¹⁵ Res., 108, 4251, doi:10.1029/2001JD001374, 2003. van der Werf, G. R., Randerson, J. T., Giglio, L., Collatz, G. J., Mu, M., Kasibhatla, P. S., Morton, D. C., DeFries, R. S., Jin, Y., and van Leeuwen, T. T.: Global fire emissions and the contribution of deforestation, savanna, forest, agricultural, and peat fires (1997–2009), Atmos. Chem. Phys., 10, 11707–11735, doi:10.5194/acp-10-11707-2010, 2010.
- ²⁰ Wang, T., Nie, W., Gao, J., Xue, L. K., Gao, X. M., Wang, X. F., Qiu, J., Poon, C. N., Meinardi, S., Blake, D., Wang, S. L., Ding, A. J., Chai, F. H., Zhang, Q. Z., and Wang, W. X.: Air quality during the 2008 Beijing Olympics: secondary pollutants and regional impact, Atmos. Chem. Phys., 10, 7603–7615, doi:10.5194/acp-10-7603-2010, 2010.

Wang, X. L., Feng, Y., Compo, G. P., Swail, V. R., Zwiers, F. W., Allan, R. J., and Sardesh-

- mukh, P. D.: Trends and low frequency variability of extra-tropical cyclone activity in the ensemble of twentieth century reanalysis, Clim. Dynam., 40, 2775–2800, doi:10.1007/s00382-012-1450-9, 2013.
 - Worden, H. M., Deeter, M. N., Edwards, D. P., Gille, J. C., Drummond, J. R., and Nédélec, P.: Observations of near-surface carbon monoxide from space using MOPITT multispectral re-
- trievals, J. Geophys. Res., 115, D18314, doi:10.1029/2010JD014242, 2010.
 Wotawa, G., Novelli, P. C., Trainer, M., and Granier, C.: Interannual variability of summertime CO concentrations in the Northern Hemisphere explained by boreal forest fires in North America

and Russia, Geophys. Res. Lett., 28, 4575-4578, 2001.



Yienger, J. J., Galanter, M., Holloway, T. A., Phadnis, M. J., Guttikunda, S. K., Carmichael, G. R., Moxim, W. J., and Levy II, H.: The episodic nature of air pollution transport from Asia to North America, J. Geophys. Res., 105, 26931–26945, doi:10.1029/2000JD900309, 2000.

Yu, R., Xu, Y., Zhou, T., and Li, J.: Relation between rainfall duration and diurnal variation

⁵ in the warm season precipitation over central eastern China, Geophys. Res. Lett., 34, Li3703,doi:10.1029/2007GL030315, 2007.

Yue, X. and Wang, H.: The springtime North Asia cyclone activity index and the Southern Annular Mode, Adv. Atmos. Sci., 25, 673–679, 2008.

Yurganov, L. N., McMillan, W. W., Dzhola, A. V., Grechko, E. I., Jones, N. B., and van

- derWerf, G.: Global AIRS and MOPITT CO measurements: validation, comparison, and links to biomass burning variations and carbon cycle, J. Geophys. Res., 113, D09301, doi:10.1029/2007JD009229, 2008.
 - Zhang, L., Jacob, D. J., Bowman, K. W., Logan, J. A., Turquety, S., Hudman, R. C., Li, Q. B., Beer, R., Worden, H. M., Worden, J. R., Rinsland, C. P., Kulawik, S. S., Lampel, M. C.,
- Shephard, M. W., Fisher, B. M., Eldering, A., and Avery, M. A.: Ozone-CO correlations determined by the TES satellite instrument in continental outflow regions, Geophys. Res. Lett., 33, L18804, doi:10.1029/2006GL026399, 2006.
 - Zhang, Q., Streets, D. G., Carmichael, G. R., He, K. B., Huo, H., Kannari, A., Klimont, Z., Park, I. S., Reddy, S., Fu, J. S., Chen, D., Duan, L., Lei, Y., Wang, L. T., and Yao, Z. L.: Asian
- 20 emissions in 2006 for the NASA INTEX-B mission, Atmos. Chem. Phys., 9, 5131–5153, doi:10.5194/acp-9-5131-2009, 2009.
 - Zhao, C., Wang, W., Yang, Y., Fu, R., Cunnold, D., and Choi, Y.: Impact of East Asian summer monsoon on the air quality over China: view from space, J. Geophys. Res., 115, D09301, doi:10.1029/2009JD012745, 2010.
- ²⁵ Zhao, T. L., Gong, S. L., Zhang, X. Y., and Jaffe, D. A.: Asian dust storm influence on North American ambient PM levels: observational evidence and controlling factors, Atmos. Chem. Phys., 8, 2717–2728, doi:10.5194/acp-8-2717-2008, 2008.

Zhou, D., Ding, A., Mao, H., Fu, C., Wang, T., Chan, L. Y., Ding, K., Zhang, Y., Liu, J., Lu, A., and Hao, N.: Impacts of the East Asian monsoon on lower tropospheric ozone over coastal

³⁰ South China, Environ. Res. Lett., 8, 044011, doi:10.1088/1748-9326/8/4/044011, 2013.



Table 1. Characterization of the three cases.

Case	2003	2004	2005		
Date	6 June 2003	18 March 2004	10 April 2005		
Maximum CO (ppbv) in MOZAIC profiles in FT	~ 550	~ 500	~ 300		
CO peak height (hPa) in MOZAIC profiles in FT	500–300	750–550	350–250		
Maximum CO (ppbv) in MOPITT images	300–400	200–250	150–200		
CO peak height (hPa) in MOPITT images	650–300	750–500	400–250		
Peak CO area in MOPITT images	35–55° N 125–145° E	20–32° N 125–135° E	32–37° N 130–140° E		
Major CO sources	Large fires near Lake Baikal in Russia	Fires in the Indochina peninsula, anthropogenic emissions in the North China Plain	Fires in the Indochina peninsula, anthropogenic emissions in the North China Plain and the Sichuan basin		
Vertical transport mechanism	Frontal lifting	Convection (due to lee- side troughs), frontal lift- ing, and orographic lifting	Convection, frontal lifting		
Outflow	West coast of Canada	West coast of the United States	West coast of Canada		

ACPD 14, 28019-28077, 2014 **Uplifting of carbon** monoxide to the free troposphere in East Asia K. Ding et al. Title Page Abstract Introduction References Tables Figures 4 Back Close Full Screen / Esc **Printer-friendly Version** Interactive Discussion $(\mathbf{\hat{H}})$

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			Occurrence				Fractional Occurrence (%)						
Season P (r	Pressure		CO Range (ppbv)										
	(hPa)	0–100	100–200	200-300	300-400	> 400	All	0-100	100–200	200-300	300-400	> 400	All
All	400-200	67	354	93	19	6	539	12	66	17	4	1	100
	600-400	36	359	69	15	8	487	7	74	14	3	2	100
	850-600	17	180	150	31	6	384	4	47	39	8	2	100
	Surface-850	4	60	142	83	88	377	1	16	38	22	23	100
Spring	400-200	11	96	28	12	2	149	7	64	19	8	1	100
	600-400	1	101	29	9	3	143	1	71	20	6	2	100
	850-600	0	38	55	20	5	118	0	32	47	17	4	100
	Surface-850	0	14	44	27	29	114	0	12	39	24	25	100
Summer	400–200	14	132	41	4	4	195	7	68	21	2	2	100
	600-400	13	138	22	1	2	176	7	78	13	1	1	100
	850-600	14	80	50	6	0	150	9	53	33	4	0	100
	Surface-850	4	32	48	30	36	150	3	21	32	20	24	100
Fall	400-200	30	61	15	2	0	108	28	56	14	2	0	100
	600-400	20	50	11	2	1	84	24	60	13	2	1	100
	850-600	3	30	17	3	0	53	6	57	32	6	0	100
	Surface-850	0	10	20	12	11	53	0	19	38	23	21	100
Winter	400–200	12	65	9	1	0	87	14	75	10	1	0	100
	600-400	2	70	7	3	2	84	2	83	8	4	2	100
	850-600	0	32	28	2	1	63	0	51	44	3	2	100
	Surface-850	0	4	30	14	12	60	0	7	50	23	20	100

Table 2. Occurrences of various CO concentration ranges at different altitudes in the MOZAIC measurements in the vicinity of Narita from 2001 to 2006.



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Figure 1. The Degree of Freedom for Signal (DFS) of the MOPITT V5 TIR/NIR data over East Asia, averaged for 2005 during **(a)** daytime and **(b)** nighttime. Locations of the airports with MOZAIC CO measurements at four cities are indicated as stars. Note that the MOZAIC CO data from Narita also include a small portion of measurements from its surrounding airports at Osaka and Nagoya.





Figure 2. Relative bias of CO profiles (in %) between MOPITT and MOZAIC data (smoothed with the MOPITT averaging kernels, see Eq. 1), also termed as "MOPITT estimate of in situ data") from 2003 to 2005 at Beijing, Narita, Shanghai, and Hong Kong for MOPITT V4 and V5 data. The number of profiles for the comparison is 18, 23, 11, and 15, respectively, at Beijing, Narita, Shanghai, and Hong Kong. The error bars indicate the interquartile range of the mean.













Figure 4. MOPITT CO mixing ratio (ppbv, in color) (a) on 6 June 2003 at 500 hPa, (b) on 18 March 2004 at 700 hPa, and (c) on 10 April 2005 at 300 hPa. All are overlaid with horizontal winds (in arrows) at the same altitude. In each subfigure, the red dot indicates the location where the MOZAIC measurement was available. The box indicates an area over which mean MOPITT CO profile was taken and displayed in Fig. 5. The two blue dashed lines define the longitudinal zone, over which the CO abundances were averaged and shown in Fig. 6. The solid blue bars in (a) and (c) indicate the locations where particles were released and backward trajectories were simulated using FLEXPART (see text for detail).





Figure 5. Profiles of MOPITT CO and the a priori, averaged over the corresponding boxed area in Fig. 4 on (a) 6 June 2003, (c) 18 March 2004, and (e) 10 April 2005. The corresponding monthly mean MOPITT CO profile over the same area is also shown. The corresponding MOZAIC CO profiles (at red dots in Fig. 4) on the same day are shown in (b), (d), and (f), respectively. The corresponding MOZAIC cozone and relative humidity profiles are also shown in (b), (d), and (f). Note that the smoothed MOZAIC CO profiles (MOZAIC CO(s)) were calculated using the averaging kernels and the a priori in the boxed area in each case (see Sect. 5 for discussion).



Discussion Paper **ACPD** 14, 28019-28077, 2014 Uplifting of carbon monoxide to the free troposphere in East **Discussion** Paper Asia K. Ding et al. **Title Page** Abstract Introduction **Discussion** Paper Conclusions References Figures Tables < Close Back **Discussion** Paper Full Screen / Esc **Printer-friendly Version** Interactive Discussion

Figure 6. A latitude–altitude cross section of MOPITT CO averaged between the two blue dashed lines in Fig. 4 on (a) 6 June 2003, (b) 18 March 2004, and (c) 10 April 2005. The contour lines indicate U wind speed (m s⁻¹). Vectors are for wind directions in V and W. For a better illustration, W is enlarged by a factor of 100. The pink box(es) in (a) and (c) indicate the locations where particles were released and backward trajectories were simulated using FLEXPART (see text for detail).



Figure 7. (a) Particle distribution between 6.25-10.25 km (~ 550-250 hPa) during 1-6 June 2003. The particles were released from two locations (in pink lines) around 400 hPa (also see Figs. 4a and 6a) on 6 June 2003 and backward trajectories were calculated. The contour lines are the geopotential heights at 850 hPa on 3 June 2003. A cold front and a warm front are indicated by green and red lines, respectively. (b) The same as (a), but between 0–3.5 km. The contour lines are the geopotential heights at 850 hPa on 2 June 2003. The circles, diamonds, and stars denote daily mean fire counts of 20–100, 100–300, and 300–500 per $2.5^{\circ} \times 2.5^{\circ}$ grid area, respectively, from 31 May to 6 June.





Figure 8. Vertical distribution of particles, varying with time from 1 June 2003 at 00:00 UTC to 16 June 2003 at 00:00 UTC. The particles were released from fire regions in Fig. 7b from the surface to 3 km on 1 June 2003 and forward trajectories were calculated (15 days). The forward time (in h) and date (in June) are indicated in the x axis on the bottom and the top, respectively.





Figure 9. MOPITT CO mixing ratio at 700 hPa from 11–19 March 2004, overlaid with the geopotential height at 850 hPa on 17 March 2004 in blue contour and with a front shown by brown solid line. The large and small stars denote daily mean fire counts of 100–200 and over 200 per $2.5^{\circ} \times 2.5^{\circ}$ grid area during the period, respectively. "L" and "H" indicate a low and high pressure system, respectively.







Figure 10. (a) CO, **(b)** CO from biomass burning, and **(c)** CO from the anthropogenic source on 18 March 2004 at 00:00 UTC, simulated by GEOS-Chem. The geopotential height at 700 hPa is indicated with white lines. "L" indicates a low pressure system.



Figure 11. Latitude-altitude cross sections along 130° E of (a) CO, (b) CO from biomass burning, and (c) anthropogenic CO on 18 March 2004 at 0:00 UTC, simulated by GEOS-Chem. The contour lines indicate U wind speed (m s⁻¹). Vectors are for wind directions in V and W. For a better illustration, W is enlarged by a factor of 100.

Pressure (x100hPa)

/100hPa

x100hPa

50 45 40



Figure 12. A longitude-altitude cross section of CO along 22° N on 17 March 2004 at 06:00 UTC, simulated by GEOS-Chem. The topography of the Hengduan Mountains is indicated in white.





Figure 13. MOPITT CO mixing ratio at 800 hPa from 3–10 April 2005, overlaid with the geopotential height at 850 hPa on 9 April 2005 at 00:00 UTC in blue contour and with a front in brown solid line. The large and small stars denote daily mean fire counts of 100–200 and over 200 per $2.5^{\circ} \times 2.5^{\circ}$ grid area during the period, respectively. The boxed area was identified as a major CO source region from the FLEXPART simulation (see text for detail).





Figure 14. The GEOS-Chem simulated CO (a) on 8 April 2005 in the lower troposphere (800–700 hPa), (b) on 9 April in the middle troposphere (500–400 hPa), and (c) on 10 April in the upper troposphere (300–200 hPa). The contours are the geopotential height at 850, 450, and 250 hPa, respectively.







Figure 15. The GEOS-Chem simulated fraction of CO (a) from biomass burning and (b) from the anthropogenic source on 10 April 2005 at 00:00 UTC in the middle troposphere (500–400 hPa), overlaid with the geopotential height at 450 hPa. (c) and (d) are the same as for (a) and (b), respectively, but in the upper troposphere (300–200 hPa), overlaid with the geopotential height at 250 hPa.