Dear Editor,

We are indeed grateful to the reviewers for their insights and critiques. The following is our point-to-point response to their questions (with line number in the manuscript if any). The comparison between the original and the revised manuscript is provided in a separate file.

With best regards,

Jane Liu

Review 1:

1. p. 28024, l. 19. The word 'contaminated' or 'contamination' (also appearing on p. 28029, l. 24; on p. 28046, l. 7; and on p. 28049, l. 12) does not seem like the best choice here; the actual point seems to be that the MOPITT vertical resolution is typically quite coarse. This is typical for satellite remote sensing products and is well understood by most users of MOPITT data. MOPITT and other CO-measuring satellite instruments cannot directly measure the volume mixing ratio at a specific pressure level, but they can accurately measure average mixing ratio over a thick layer.
Thanks for the points. These points are described in Discussion (L768-711). Word "contamination" is not used.

2. p. 28025, l. 17. The meaning of ' ... a swath of 29 pixels ...' is not clear. One cross-track scan of the MOPITT instrument actually generates 29 x 4 = 116 pixels.
Thanks. "4 pixels in a row" is added (L140).

3. p. 28025, l. 21. The meaning of 'complete global coverage' is not clear, since persistently cloudy areas (such as areas of the Amazon Basin) might not be observed at all in a continuous period of 16 days. Is there a reference for this statement regarding complete global coverage?
The phrase is removed.

4. p. 28026, l. 6. In addition to the MOPITT version number, this paragraph should state which level of MOPITT data was used. Level 2 (individual retrievals) or Level 3 (gridded)?
"Level 2" is added (L158).

5. p. 28024, l. 23. Add reference to MOPITT Version 5 validation paper (Deeter, M. N., et al (2013), Validation of MOPITT Version 5 thermal-infrared, near-infrared, and multispectral carbon monoxide profile retrievals for 2000–2011, J. Geophys. Res., doi:10.1002/jgrd.50272).
This reference is added (L991) and discussed in this revision (L300-306).

6. p. 28026, l. 4. Since MOPITT V5 data are used extensively in this paper, there should be some discussion of (and reference to) the results presented in the MOPITT V5 validation paper. For example, results in that paper indicate a retrieval bias in the upper troposphere. Would that explain some of the features of the MOPITT/MOZAIC comparisons shown in Fig. 2?
Yes, Deeter et al. (2013) compared MOPITT data with the NOAA aircraft measurements over North America and data from the HIAPER Pole to Pole Observations (HIPPO) field campaign data (Wofsy et al., 2011). They found a positive bias in MOPITT V5 TIR/NIR data at 400 hPa (4%) and 200 hPa (14%). They also showed a latitude-dependent positive bias in the northern hemispherical upper troposphere in MOPITT V3 and V4 data. This study suggests an overall positive bias in the upper troposphere, agreeing with Deeter et al. (2013) in magnitude and sign, in MOPITT V5 data. We have added the discussion for Fig. 2 (L300-306).

7. p. 28026, l. 19. Important details seem to be missing in this section (and in the captions to Figures 2 and 3) concerning the method used to identify MOPITT observations corresponding to a particular MOZAIC flight. MOZAIC vertical 'profiles' are actually produced by observations made over a slant path with varying latitude and longitude. For each MOZAIC flight, was the MOPITT collocation radius (1.5 degrees) applied to a single MOZAIC lat/lon location at a specific altitude or to all of the MOZAIC lat/lon values within some altitude range? Also, are the results presented in Figures 2 and 3 sensitive to the chosen collocation radius?
The radius of 1.5 °is applied to a selected MOZAIC profile at 500 hPa so that the MOZAIC slant path can be included. Only when the entire slant path is within the radius, the MOZAIC profile is selected (L171-172). Because of adequate MOPITT samplings in the radius, the results in Figures 2 and 3 are not affected much by slight change in radius.

8. p. 28032, l. 22. The meaning of 'some degree of vertical sensitivity' is unclear. Does this statement refer to the ability to detect enhanced CO at a particular level, or to the vertical resolution?
We clarified this in this version of the manuscript. The vertical sensitivity is demonstrated through (1) the strongest CO source among the three cases was captured by the largest magnitude of CO enhancement of 200-250 ppbv from the a priori, (2) the altitude of the maximum CO enhancement was detected around the middle troposphere, in contrast to the other two cases which show the maximum in the lower-middle and upper troposphere, respectively, and (3) the elevated CO was over a broad range of altitudes as the vertical resolution of MOPITT is rather coarse, i.e., the maximum DFS is about 2.5 (Figure 1) (L348-354).

9. For all case studies, what criteria were used to determine the locations and shapes of the MOPITT boxes shown in Figure 4 determined?
The criteria are to ensure enough samplings of MOPITT measurements (no less than 30 data points) at the closest upwind direction of MOZAIC measurements. We have explained this in the caption (L1384-1385).

10. p. 28046, l. 5. Suggest replacing 'smooth MOZAIC profiles' with 'averaging kernel smoothed MOZAIC profiles.'
Changed (L783).

11. p. 28047, l. 16. 'MOPITT satellite' should be 'MOPITT satellite instrument'.
Changed (L810).

12. p. 28048, l. 17. 'frontal activates' should be 'frontal activity'
Corrected (L842).

13. p. 28049, l. 19. The last sentence of the Conclusion is unclear and seems to imply a bias in the MOPITT data. The statement 'MOPITT substantially underestimates CO in high CO episodes' really seems to be referring to the fact that remote sensing instruments like MOPITT cannot resolve sharp peaks in the CO profile. This is an issue of vertical resolution and does not imply a bias.
This sentence is removed.

14. p. 28066, Fig. 4. In addition to showing the location of the airport in each panel as an indicator of the location of the MOZAIC profile (i.e., the red dots), the figure should show a series of points indicating the actual latitude and longitude of the MOZAIC data at various altitudes or pressures (e.g., at 1 km or 100 hPa intervals).
Thanks for the suggestion. The geographic locations of MOZAIC data at 900, 600 and 300 hPa are now added in Figure 4.

15. p. 28073, Fig. 11. In this figure, why does latitude decrease from left to right? This will certainly confuse most readers.
Figure 11 and Figure 6 are re-plotted with increasing latitude from left to right.

Review 2:
One thing that wasn't clear to me is how frequent are these episodes of enhanced vertical transport. The authors identify just 3 cases across several years. Are these episodes hard to spot in the sparse data? Very infrequent? If the latter, do they add up to an important contribution regionally? world-wide? Does the model see more of them? Or are they a case study to more easily learn about vertical transport? It would be very useful to read more background.

Thanks for the points. More background information is now provided and discussed in Sect. 5.1 (L663-688). The frequency of high CO episodes observed by MOZAIC around Narita was summarized in original Table 2. The frequency of high CO episodes appeared in MOPITT and GEOS-Chem data is provided in a new Table 3. According to these data, it is likely that the high CO episodes shown in this study occurred 2-4 times every 100 days for each lower, middle and upper troposphere over the Sea of Japan and the East China Sea. GEOS-Chem does not see more high CO episodes in the free troposphere than MOZAIC or MOPITT. Overall, MOZAIC observed slightly more transport of high CO to the upper troposphere, while GEOS-Chem simulates more transport of CO (with lower abundances) to the middle and lower troposphere. According to Tables 2 and 3, air mass with 200-300 ppbv CO is transported to 400-200 hPa at a frequency of 10-20 times per 100 days, or approximately once a week. This can have significant impacts on the air quality downwind. The transport mechanisms and CO source contributions revealed in this study can also be applicable for these CO episodes, even at lower CO abundances or lower altitudes (L663-688).

In general, I think the sections should be shortened and made more concise, especially given the lack of quantitative information. The authors are strongly encouraged to add quantitative information.
The contents in the original Sect.5 (Discussion) are substantially removed. We have added more quantitative information in Discussion and in Table 3.

p28025, l 5-10. It's a bit confusing to read about demonstrating MOPITT's vertical sensitivity, since I thought it was MOPITT's vertical sensitivity that was being used to evaluate the vertical transport, not the other way around. It would be great to be clear about that here already.
MOPITT data are analyzed in two ways in this study. First, the vertical sensitivity of MOPITT is evaluated with the coincident MOZAIC data (see Figures 1, 2 and 3) and further illustrated with in the high CO episodes in comparison with the MOZAIC data (see Figure 5). Second, the vertical variation in CO captured by MOPITT is used to diagnose vertical transport of CO (Figure 6) (L125-131).

p28027, l 17-19. It would be great to put the list of parameters at the beginning of this subsection, otherwise, it's a bit unclear what "analyses" include Section 2.5 Are the met data described in 2.3 used only to drive FLEXPART? Perhaps it's better to combine those two sections then, similarly to how GMAO met fields are in the same section as GEOS-Chem Section 2.6.
The list of parameters is moved to the beginning of this section. We added "In addition to driving FLEXPART, the FNL data are used to analyze the meteorological conditions including the surface pressure, wind fields, and development of a cyclone." (L208-210). We moved this section right ahead of

the section on FLEXPART.

Is the full chemistry version of GEOS-Chem being used as implied or is it just tagged CO/single tracer simulation?
GEOS-Chem was used in the full chemistry mode. This information is added into the text (L234).

p.28030, l20-21. I don't understand the concept of "difference between averaging kernels", is it its diagonals?
The difference is the averaging kernel for each retrieved pressure level between version 5 and version 4.

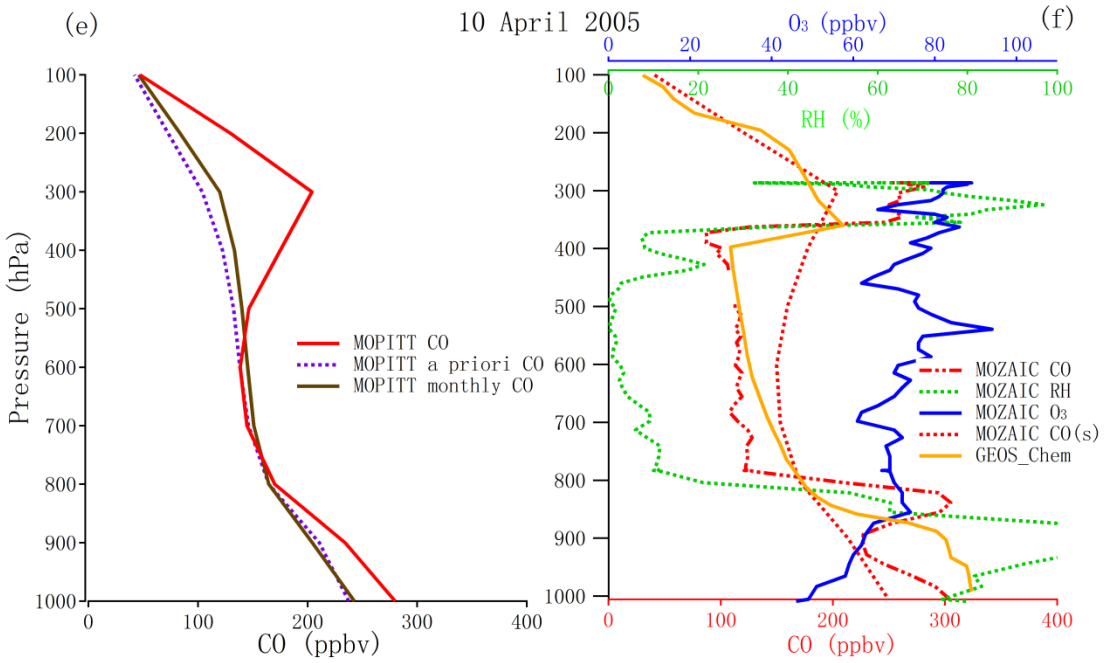
Section 4.1 It's an interesting reconstruction of the history of a pollution plume, but I'm not sure how much of this was not known before. It's not clear what's new here. Is it that pollution is transported via WCB? That it is lofted 9 km? That it reaches Canada? That MOPITT and models agree? None of this sounds new, so it would be helpful if the authors stated more explicitly what new insights they have gained here. I'm also curious what does GEOS-Chem say about this case study.
Thanks for the comments. New insights gained from this case are now discussed at the end of Sect. 4.1 (L417-422). More detail on GEOS-Chem simulation for this case is provided in Sect. 5.5 (L743-745).

p28036, l2. "This source is confirmed in the GEOS-Chem simulation". Are the authors really relying more on GEOS-Chem than MOPITT to confirm cases of vertical transport of pollution? Shouldn't it be the other way around? especially at 700hPa. Naturally, it would be good to have independent data set here, but I guess MOZAIC was not available? It would be good if the sections were a bit more parallel (each commenting on the skill of both models and availability and quality of both data sets) Is Figure 12 necessary?
Thanks for the points. This study aims for solving puzzles of vertical transport of CO, using observations from MOZAIC and MOPITT and simulations from FLEXPART and GEOS-Chem, as well as other data. All the data and simulations were integrated to examine if they support or complement each other without particular preference to one dataset or simulation. In this case, elevated CO plumes around 700 hPa is captured in MOZAIC and MOPITT data and GEOS-Chem simulation (Figures 4, 5, 6), while in the upper troposphere, the elevated CO is missed in MOZAIC data but captured by GEOS-Chem (Figure 11), and weakly detected by MOPITT (Figure 6b). Because both MOPITT and MOZAIC data only show the sum of CO from all sources so we used fire data and GEOS-Chem simulation to separate different sources. We have replaced the word "confirm" with "recognized" to express ourselves more clearly (L458). We have made the sections more balanced with the observations and simulations. We have commented on the skill of

both models in Sect. 5.4 (L736-754) and availability and quality of MOPITT data in Sect.5.5 (L757-803).
Figure 12 is necessary because it illustrates an important finding in this study (see Sect. 4.2).

p28038, l 1-3 How was this somewhat random time frame chosen? I'm sure there was a good reason, but it's not clear from the text what it was.
Thanks for pointing this out. We did backward trajectories first from the boxed area in Figure 4b, and these trajectories indicate the most particles mainly came from the large fire regions starting from 11 March 2004. We have explained this now in the text (L510-511).

p28038, l 21. It's a bit difficult to believe that different altitude of this plume as identified by MOPITT is an indication of MOPITT's ability to resolve vertical structure. While this could be true given how good MOPITT instrument is in general, the earlier section relied more on GEOS-Chem than MOPITT to even identify the plume, so here I am wondering if we should take MOPITT at face value or wonder what GEOS-Chem is showing in this case.
As mentioned earlier, we did not particularly rely on one dataset or simulation. Instead, we put all the data and simulations together to examine if they support or complement each other. We did examine the GEOS-Chem profile over the boxed area in Figure 4c. GEOS-Chem also shows elevated CO plumes around 400-200 hPa (shown below), which supports the MOZAIC and MOPITT observations. The GEOS-Chem profile was not included in Figure 5f in order to keep it simple and focused, while the signal of the CO plume was illustrated and discussed in Figures 14 and 15.



Section 5 Discussion. It's hard to tell what is the exciting findings. Is it that there was high CO documented? Is it that topography affects vertical CO transport? Is either new and/or surprising? Please tell the reader so. It all seems intuitive and the section is purely qualitative, so it's not clear that anything new is being reported. It would also be helpful to have quantitative information. For example, how does topography affect CO transport, does it have to be a mountain region? only in the north/east/west/south? does any of this or could any of this vary with seasons? It's hard to draw conclusions from individual case studies.

Thanks for the points. The background information is provided in Sect. 5.1 (L650-688). The topography effects are discussed in Sect. 4.2 (L492-496). This section is rewritten to address the reviewer questions. The main contributions of this study include (1) observing rare high CO episodes in the free troposphere in East Asia, (2) identifying distinct transport mechanisms, pathways, and CO sources for these episodes, (3) supporting and extending a proposed mechanism of the leeside troughs over the Indochina peninsula in promoting vertical transport of pollutants, and (4) analyzing MOPITT data from perspective of its vertical sensitivity at synoptic scale (L633-803).

p28044, l28-29 It's not clear why the statistical analysis couldn't be done here already. Authors should consider doing at least preliminary work on that.

In the previous manuscript, we suggested this work as a direction for further studies. This suggested work requires long-term analysis, or at least longer than 5 year. In order to provide accurate statistics, careful examinations of each WCB for the period are needed. This is a large undertaking itself and also beyond the scope of this study. It does not affect the conclusions of this study.

p28045-28046. These whole pages (and the remainder of that section) can be deleted, especially the first paragraph. It's just repeating background information.

This part is largely shortened. The application of MOPITT data is one of the major focuses of this study so some discussion on this perspective is desirable. We now emphasize more on new insights gained from this study (L755-803).

p28048, l17 replace "interplaying" with "interacting"?

Replaced (L840).

Figures: figure captions and labels are a bit too small to be readable, especially on figures 1-7, figure 11.

Figure captions and labels are enlarged for these figures.

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

K. Ding[1,6], J. Liu[1,2,6,*], A. Ding[1,6], Q. Liu[1,6], T. L. Zhao[3], J. Shi[4], Y. Han[1], H. Wang[5], F. Jiang[5]

[1]School of Atmospheric Sciences, Nanjing University, Nanjing, Jiangsu, 210093, China

[2]University of Toronto, Toronto, Ontario, M5S 3G3, Canada

[3]Nanjing University of Information Science & Technology, Nanjing, Jiangsu, 210044, China

[4]Institute of Remote Sensing Applications, Chinese Academy of Sciences, Beijing, 100101, China

[5]International Institute for Earth System Sciences, Nanjing University, Nanjing, Jiangsu, 210093, China

[6]Collaborative Innovation Center of Climate Change, Jiangsu, 210093, China

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[*]Corresponding author

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~~.~~ around the world. It can also influence global climate due to the longer lifetime of CO at higher altitudes. In this study, three cases of high CO episodes in the East ~~Asia~~China Sea and the Sea of Japan 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~~are observed in MOZAIC data in the free troposphere during these episodes. These are among the highest CO abundances documented at these altitudes~~.~~, likely occurring 2-4% in time in the respective altitudes over the region. Correspondingly, elevated CO ~~was~~is 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 transport 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, originated 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 in the free troposphere 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 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 (CO_2) 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 et al., 2004). Uplifting airmass from the surface to FT generally takes place by three 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 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 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 extratropical 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 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 populated 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 understanding 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., 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.~~). 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~~influenced by CO at other levels and thus MOPITT vertical resolution is coarse, generally having only 2-3 pieces of independent information vertically in the troposphere. MOPITT's vertical sensitivity ~~becomes a concern~~was an issue 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. 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 hPa 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. 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; 2013). 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~~ MOPITT data are analyzed in two ways. First, 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~~ with the three high CO episodes in comparison with the MOZAIC data (see Sect.4). Second, the vertical variation in CO captured by MOPITT is used to diagnose vertical transport of CO (see Sect. 4). Discussion on the three cases is synthesized in Sect.5 and the major conclusions are provided in Sect.6.

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~~,~~ (4 pixels in a row), each ~~of them~~pixel being ~~22km x 22km~~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 achieve a near-complete global coverage ~~and 16 days for a complete global coverage~~(Edwards et al., 1999) if assuming no blocks from clouds.

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 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~~). ~~; 2013). 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, 2013).

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~~.~~ with the level 2 data. Then, V5 level 2 data were used throughout the case studies, in

which MOPITT data were gridded horizontally into 0.25 °latitude ~~x~~× 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 automatic 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 ~~4h period. Each~~4 h period. The radius of 1.5 °was applied to selected MOZAIC profiles at 500 hPa and the MOZAIC slant path was included in the radius. 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 $\hat{\mathbf{x}}^{MOZAIC}$ is derived by (Rogers, 2000)

$$\hat{\mathbf{x}}^{MOZAIC} = \mathbf{x}_a^{MOPITT} + \mathbf{A}\left(\mathbf{x}^{MOZAIC} - \mathbf{x}_a^{MOPITT}\right) \quad (1)$$

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

MOZAIC CO

_profile, which has been mapped to the MOPITT pressure grid. The quantity $\mathbf{x}_{\mathbf{a}}^{MOPITT}$ 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 ~~250hPa~~ 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 NCEP FNL meteorological data~~

~~The Final (FNL) global tropospheric analyses are on 1 °by 1 °grids every 6h (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 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.~~

~~2.4 The~~

2.3 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 approach to exploit the strong emission of midinfrared radiation from fires allowing subpixel 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).

2.4 NCEP FNL meteorological data

The NCEP Final (FNL) global tropospheric analyses are on 1 °by 1 °grids every 6h (http://rda.ucar.edu/datasets/ds083.2/). Parameters in FNL 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, available on the surface, at 26 levels from 1000 to 10 hPa, the tropopause, the boundary layer, and a few others. In addition to driving FLEXPART (see Sect. 2.5), the FNL data are used to analyze the meteorological conditions including the surface pressure, wind fields, and development of a cyclone. The data are generated from the Global Data Assimilation System (GDAS).

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 (latitude, longitude, and altitude) at a time. Backward or forward trajectories of the particles 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-NOx-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 and executed in the full chemistry mode, which is driven by GEOS meteorology with temporal resolution of 6h (3h for surface

meteorological variables), with a horizontal resolution of 2 °latitude by 2.5 °longitude and 47 vertical levels, including ~35 levels in the troposphere from 1000 to ~~100hPa~~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~~contribution of CO from other levels to the retrieved level 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). Figure 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. The annual mean DFS is 1.65, 1.51, 1.60, and 1.64, respectively, in an area of 1 °x1 °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.

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~~in the surface and lower troposphere and as 0.10 ~~for~~in the upper troposphere (not shown).

Figure 2 shows the relative bias between MOPITT and the smoothed MOZAIC ($\hat{\mathbf{x}}^{MOZAIC}$) 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 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. Deeter et al. (2013) compared MOPITT data with the NOAA aircraft measurements over North America and data from the HIAPER Pole to Pole Observations

(HIPPO) field campaign data (Wofsy et al., 2011). They found a positive bias in MOPITT V5 TIR/NIR data at 400 hPa (4%) and 200 hPa (14%). They also showed a latitude-dependent positive bias in the northern hemispherical upper troposphere in MOPITT V3 and V4 data. This study suggests an overall positive bias, agreeing with Deeter et al. (2013) in magnitude and sign, in MOPITT V5 data for the upper troposphere. As a comparison, we also validated MOPITT data in other cities 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).~~ 3. 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, respectively (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).

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 three cases, high CO concentrations up to 300-500 ppbv are observed by MOZAIC in the free troposphere from 750 to 350 hPa. ~~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 layer at the west coast of the United States and Canada.

4.1 Case study I: 6 June 2003

On 6 June 2003, a large area (~ 400 km ~~x~~× 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~~650-300 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.~~ capability of capturing pollution episodes with some degree of vertical sensitivity. The vertical sensitivity is demonstrated through (1) the strongest CO source among the three cases was captured by the largest magnitude of CO enhancement of 200-250 ppbv from the a priori, (2) the altitude of the maximum CO enhancement was detected around the middle troposphere, in contrast to the other two cases which show the maximum in the lower-middle and upper troposphere, respectively, and (3) the elevated CO was over a broad range of altitudes as the vertical resolution of MOPITT is rather coarse, i.e., the maximum DFS is about 2.5 (Figure 1). 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 measurement (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-~~50N. 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.~~50 °N.

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, 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~~°x~~ × 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~~60 °N, 0-3 km, following ~~Lavoue~~Lavou é 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). The altitudes with high particle concentrations agree ~~remarkably~~ well the MOPITT data between ~~500~~650-350 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 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 analyzed 320 MOZAIC flight routes from~~

Our analyses are consistent with Nédélec et al. (2005), who 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~~Nédélec et al. (2005) also attribute the high CO at these altitudes to front lifting of CO from large forest fires near Lake Baikal. Furthermore, this analysis provides a detailed description on the CO transport pathways (Figs. 7 and 8). We found that this case, caused by the strongest CO source among the three cases, show the largest horizontal area with CO plumes (Fig. 5), the widest vertical CO buildup with strongest abundances (Fig. 7), and the biggest enhancement of 200-250 ppbv from the a priori (Fig. 6).

4.2 Case study II: 18 March 2004

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 ~~it's~~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 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~~March 2004 at 700 hPa over northern Indochina peninsula (Fig. 9). ~~This source is confirmed in~~In the GEOS-Chem simulation (Fig. 10b~~).~~), this source is also recognized. In addition, the anthropogenic source concentrated over the North China Plain (NCP) (approximately 30-40 °N, 110-125 °E) is identified as another source of high CO ~~in the GEOS-Chem simulation~~ (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 ~ 750 hPa. 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.~~2m~~2 m s^{-1} in FNL data around ~~at~~ this level (not shown). The ECMWF (European Centre for Medium-Range Weather Forecasts) 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 lifted CO was with even higher 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 ~~700hPa~~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-c).

The Hengduan Mountains run mainly north to south, with elevations ranging from 1300 to 6000 metres. This topography provides a favorable condition for the formation of the leeside troughs if meteorology is satisfied. Such troughs promote vertical transport of CO on the west flank of the Mountain (in the second and fourth processes), while the orographic lifting occurred on the east flank of the Mountains (in the first process).

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 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~~As the backward trajectories starting from the boxed area in Figure 4b indicated the most particles came from the large fire regions starting from 11 March 2004, air particles were released in FLEXPART over the ~~large fire~~ regions from the surface to 1 km on 11 March 2004~~. FLEXPART was used~~, and forward trajectories were simulated to ~~simulate transport of~~track down 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

555 southerly outflow.

556 The transport mechanism of biomass burning by the fourth processes was first
557 proposed by Lin et al. (2009) who found ~~a significant role~~ that the leeside troughs above
558 the Indochina peninsula play a significant role in uplifting of ozone ~~from biomass~~
559 ~~burning~~there as there is lack of tropical deep convection to explain such strong
560 convection. In this study, we even found these leeside troughs promote vertical transport
561 of CO to the upper troposphere (the second process, Fig. 12). It is the interplay of the
562 leeside troughs and the cyclone in the northeast of China which formed a front system
563 that transported CO from the Indochina peninsula upward.
564

565 ### 4.3 Case ~~Study~~study III: 10 April 2005

566 In this case, MOPITT observed high CO of ~ 250 ppbv at 300 hPa near the east coast
567 of Japan on 10 April 2005 (Fig. 4c). To compare MOPITT and MOZAIC data, the mean
568 MOPITT profile was taken over a boxed area (in Fig. 4c) upwind of the MOZAIC
569 measurement. The MOPITT vertical profile clearly shows a CO peak around 300 hPa,
570 where it departs from the MOPITT monthly mean (Fig. 5e). Comparing with cases 2003
571 and 2004, MOPITT CO peaked at higher altitudes, illustrating some MOPITT vertical
572 sensitivity even at these altitudes. In Fig. 5f, a sharp peak of 300 ppbv in MOZAIC CO
573 is shown around 350 hPa. The profile of relative humidity ~~followed~~follows closely that of
574 CO, with
575 values up to 90-100 % around ~~300~~350 hPa, implying that the elevated CO was lifted to
576 this level from the lower troposphere by a cyclone system along its WCB. However, the
577 MOZAIC ozone profile varies ~~strongly~~differently from the CO and relative humidity

profiles. This ~~was~~is found ~~to be~~ due to a strong stratospheric intrusion introduced by the cyclone. HYSPLIT simulations suggest that a large amount of ~~airmass~~air mass plunged around 4 ~~June~~April 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 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~~Figs. 14b and ~~c~~14c 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~~images 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 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~~Figs. 15a and ~~c~~15c) and from the anthropogenic source (~~Fig~~Figs. 15b and ~~d~~15d) in the middle and upper troposphere, respectively, overlaid with the geopotential height at 450 hPa (~~Fig~~Figs. 15a and ~~b~~15b) and 250 hPa (~~Fig~~Figs. 15c and ~~d~~15d), 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 topography's role was noticed by Liu et al. (2003), who found a ring of convergence around 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 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~~middle troposphere 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 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 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 °N, 100-120 °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~~20 m s^{-1} at 750 hPa, and increasing with altitude until 300-250 hPa where the maximum vertical velocity was 0.~~259~~26 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 MOPITT 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

New insights gained from this study and suggestions for future work are discussed as follows.

5.1 Observations of high CO episodes

In the three CO episodes, high CO abundances ~~above~~ 300-550 ppbv were observed ~~in aircraft~~by 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~~ (Fig 5). The elevated CO abundances 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~~Nédélec 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. For example, in all the cases, biomass burning occurred from regions 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, enhancing chances of high CO episodes in these seasons.

The ~~possibility~~frequency of occurrences of such high CO is ~~further~~ illustrated in Table 2. As the three cases ~~o~~curred~~occurred~~ near Japan, MOZAIC data around the vicinity of Narita ~~and its surrounding areas~~ from 2001 to 2006 are summarized, showing occurrences of various CO abundance 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 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.~~ The frequency of such high CO episodes is also examined in the GEOS-Chem simulations and MOPITT observations in the vicinity of Narita (126-140 °E 30-40 °N) in 2005 (Table 3). For the GEOS-Chem simulations, a count is added to a CO range if the daily maximum CO in the area at a layer falling into that CO range. Thus, the total counts for all the CO ranges at a given layer are 365, while the counts are 281 for

MOPITT due to missing data. To minimize noise in daily MOPITT data, only when there were at least 10 data with the maximum CO values falling into a given CO range, a count is added. GEOS-Chem can simulate CO up to 400 ppbv in the upper troposphere, while the maximum CO in MOPITT data is lower so that different CO ranges are used in Table 3. Overall, MOZAIC, MOPITT and GEOS-Chem all show a high frequency of high CO (larger than 200 ppbv) at the surface, gradually shifting to a high frequency of low CO (less than 200 ppbv) at the upper troposphere. Between 400-200 hPa, CO episodes with 200-300 ppbv occurred 1.2 times every 10 days in GEOS-Chem, slightly lower than in MOPITT (1.8 times) and MOZAIC (1.7 times). Overall, MOZAIC observes 2-5% more transport of high CO to the upper troposphere than GEOS-Chem, while the latter simulates 10-20% more transport of CO (with lower abundances) to the middle and lower troposphere.

It is likely that on average, the extremely high CO episodes like the three cases (Fig. 5) occurred 2-4 times per 100 days in their respective altitudes over the East China Sea and the Sea of Japan (Tables 2 and 3). As stated, air mass with lower CO abundances of 200-300 ppbv can be transported to 400-200 hPa in a frequency of 1-2 times per 10 days (Tables 2 and 3). The frequency can be even higher in spring and summer (Table 2), approximately once a week. Significant impacts of such transport can be expected on the air quality downwind and on the global climate. The transport mechanisms and CO source contributions revealed in this study can also be applicable for these CO episodes, even with lower CO abundances or at lower altitudes.

5.2 The role of topography

East Asia's topography varies significantly across its vast width, increasing from east to west, with a variety of terrains. This study found that topography there affected the three cases in different ways. 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 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). Lin et al. (2009) mainly aimed at vertical transport of pollutions to the lower and middle troposphere. Extending from Lin et al. (2009), this study found it possible to explain pollution transport to the upper troposphere using such a mechanism.

5.3 The implications of WCB trends on uplifting of CO

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 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. Although the functions and characteristics of WCBs have been recognized by earlier studies, this study provides some details unique

for the three cases. In case 2004, we found that 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. This case also appeared the most southerly among the three, leading to a most southerly outflow. In case 2005, we found downwelling of stratospheric clean air on the back side of cyclones. We also found that anthropogenic CO from two regions and CO from biomass burning dominated different parts of a WCB. The source allocation was sensitive to the location of the front. Comparing cases 2004 and 2005, we found that when large CO source coincided with the strongest part of a WCB, uplifting of CO to the upper troposphere became more possible. ~~the frontal cyclones in~~ 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 in two regions in spring and summer: 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; 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.~~

~~GEOS-Chem and FLEXPART simulations reveal different CO signatures from biomass burning and anthropogenic sources in the high CO episodes, 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, 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 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.~~investigation. It would be helpful to conduct statistical analysis of the CO source distribution along WCBs in East Asia in the future.

5.4 Comparison between observations from aircraft and satellite and model simulations

Pollution transport can be tracked computationally with Euler and Lagrangian approaches, as represented by GEOS-Chem and FLEXPART models, respectively. 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~~MOPITT~~ satellite data ~~can~~(Figs. 10, 11, and 14). This study found that GEOS-Chem simulated the observed aircraft and satellite CO well in cases 2004 and 2005 but cannot fully reproduce the elevated CO in MOZAIC data in case 2003. The CO plume is simulated in lower mixing ratios at lower altitudes than in the MOZAIC data. The plume also appeared further ~~extend MOZAIC CO observations~~ north. This is possibly due to an underestimated fire inventory or conservative parameterizations in simulating large forest fires or both in GEOS-Chem. 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 FLEXPART and satellite and aircraft observations can be found in various places on small scales.~~larger areas and over wider altitude ranges. High CO concentrations were observed by MOPITT over thousand kilometers horizontally and a few kilometers vertically~~ FLEXPART is good at simulating strong sources, while weak sources sometimes are omitted.

5.5 Applications of MOPITT data

We analyzed MOPITT data from two aspects: vertical sensitivity on the synoptic scale. ~~Such variations of CO~~Both are ~~usually diluted~~challenging and have not been studied adequately.

Large gaps due to clouds and the limited MOPITT swath make application of MOPITT on ~~longer time scales. Earlier applications~~the synoptic scale difficult. Thus application of MOPITT ~~CO~~ data over East Asia were mostly focused on monthly or seasonal ~~CO variations~~scales (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~~This study shows that even with large gaps, daily MOPITT data can capture vertical disturbances of CO on the synoptic scale, which are usually diluted on longer time scales. This study also suggests the importance of filling the gaps with other satellite data or in designing 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 chemical weather forecast in the future (Lawrence et al., 2003)~~satellite instruments, for the purpose of detecting such variation over large area on the regional and global scales.

~~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~~Typically for satellite remote sensing products, the MOPITT retrieval at a specific pressure level is influenced by CO from other levels ~~are within 20 %,~~

~~with the lowest~~and thus its retrieval at that pressure level can be 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~~ . However, MOPITT can more accurately measure average CO mixing ratio over a ~~better understanding of the atmospheric chemistry,~~thick layer. This results in a coarse vertical resolution and 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~~ This study addressed the MOPITT ~~indeed has some~~ vertical sensitivity~~.~~ with newly ~~Kar et al. (2004) found Asian summer monsoon plumes in~~ MOPITT ~~CO~~V5 data ~~as a strong enhancement of CO~~and found enhanced vertical sensitivity in V5 data in the free troposphere, even the upper troposphere ~~over India and southern China.~~, in addition to in the boundary layer emphasized by Worden et al. (2010) and Deeter et al. (~~2004)~~2012). The enhanced DFSs and the averaging kernels in V5 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~~by Worden 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.~~., (2010) and Deeter et al. (2012) are supported (Fig. 3 and Sect.3).

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 ° 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~~averaging kernel smoothed 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 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.~~ (2003). 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. Overall, this study found: (1) MOPITT can differentiate the magnitude of different CO plumes (Figs. 5 and 6), (2) MOPITT can distinguish elevated CO in the lower, middle, and upper troposphere (Figs. 5 and 6), (3) the shape of CO plumes in vertical direction matches with simulations of GEOS-Chem and FLEXPART, sometimes remarkably well (Fig.6), and (4) there is more vertical structure in CO in new V5 than in earlier versions of MOPITT data (Fig. 5).

It is the relative variations in MOPITT CO data that help diagnose of CO transport vertically or horizontally. This study suggests using MOPITT data quantitatively with caution, especially at altitudes with high CO plumes, as illustrated that the magnitude of elevated CO in MOPITT retrievals can be lower than in the MOZAIC data at altitudes where CO peaked (Fig. 5). Therefore, the vertical variation of CO, even enhanced in V5, is still much smoothed in MOPITT data. MOPITT can distinguish elevated CO in different layers of the free troposphere, yet sometimes cannot specify the exact altitude of elevated CO shown in the MOZAIC measurements (Fig. 5). One limitation for MOPITT's application of vertical transport is the complication 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 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 FLEXPART and satellite and aircraft observations can be found in various places on small scales.~~

6 Conclusions

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 airborne MOZAIC ~~aircraft~~ and ~~the~~spaceborne MOPITT ~~satellite~~instruments 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~~following conclusions.

~~conclusions as follows.~~

~~1.~~ In the three CO episodes, high CO abundances of 300-550 ppbv are observed in aircraft MOZAIC data in the free troposphere~~.~~ over the East China Sea and the Sea of Japan. These are among the highest CO abundances ever documented at these altitudes. The three cases occurred in the

1. seasons and at locations where meteorological and CO source conditions are favorable for such episodes. It is likely that on average, such high episodes occur 2-4 times per 100 days at the respective altitudes in the region. CO episodes in lower altitudes and with lower abundance occur more frequently in the region, about 1-4 times every 10 days with 200-300 ppbv in 600-400 hPa.

~~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~~in Indochina was transported through two separate pathways, leading to two distinct CO enhancements around 700 hPa and 300 hPa. In case 2005, along

2. 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.

~~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~~supported by this study in explaining CO transport to the middle troposphere and further extended ~~in this study.~~for CO transport to the upper troposphere. Strong convection

3. from the Sichuan basin also plays an important role in vertically transporting anthropogenic CO~~ to the free troposphere.~~. The topography ~~interplaying~~interacting with frontal ~~activates~~activities 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.

6. The MOPITT's vertical sensitivity is found to be enhanced in its new V5 NIR/TIR data~~.~~ in the free troposphere, even in the upper troposphere. The daytime V5 data can detect synoptic ~~disturbance~~disturbances of weather systems on horizontal 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~~smoothed by CO from other levels, the MOPITT retrievals usually underestimate elevated CO peaks ~~during the~~at altitudes with high CO ~~episodes~~plumes. 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~~On average, MOPITT slightly ~~overestimats~~overestimates the background CO in the upper troposphere.

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