

Reply to Meiyun Lin's comments:

1. "About Lines 15-20, you might also want to cite Lin et al (2012, JGR) and Lin et al (2014, Nature Geoscience) regarding Asian pollution influence on downwind regions. In particular, Lin et al (2014) demonstrated for the first time the role of changing atmospheric circulation in contributing to variability of Asian ozone pollution reaching Hawaii."

Reply - We have included Lin et al. (2012, 2014) in the revised text: "Elevated O₃ levels not only lead to degradation of local and regional air quality (Wang et al., 2006; Wang et al. 2008; Lin et al., 2008), but also have significant implications for chemical environment and air quality in downwind regions (e.g., Hudman et al., 2004; Lin et al., 2012, 2014)."

2. "A number of papers have also examined the role of biomass burning and Asian monsoon on seasonal variations of ozone over central eastern China, and thus should be included in your literature review."

Reply - We have added in the literature review: "Studies of the sources and variability of LT O₃ over China have also emphasized the roles of biomass burning emissions (Liu et al., 1999; Fu et al., 2007; Lin et al., 2009), biogenic emissions (Fu et al., 2007), and the impact of the Asian monsoon system (Liu et al., 2002; Lin et al., 2009)."

Reply to Referee #1's comments:

Thank Referee #1 for helpful comments.

1. "Section 3, Page 32592, lines 4-5: Awkward sentence. Change to 'Measurements were made by both platforms on four days: 1, 3, 11, and 15 May.' "

Reply - Changed accordingly.

2. "Section 3, Page 32594: Lines 1-5: Please be specific how "enhanced" is defined. Was there a threshold?"

Reply - We have revised the text to "The O₃ concentrations (~65 ppbv) at ~1.5 km in the average profile reflects O₃ enhancements (>70 ppbv) frequently observed in individual profiles during April-May, 2005."

3. "Section 4, Page 32596, Lines 1-5: Is there another meteorological scale that could be missing and is important? Is that what is implied here?"

Reply - The original text was not clear. We have revised it to: "The distribution and variability of model relative humidity (RH) are generally similar to those observed by ozonesondes (not shown), suggesting that convective transport and large-scale ascending and descending motions in the study region are reasonably represented in the GEOS-4 and GEOS-5 meteorology."

4. "Section 5.1, Page 32598, line 8 and others: The word "suppressed" is not a good choice here. If the emissions were completely turned off in the model, as I suspect they were, then do not use the word "suppressed" throughout this paragraph. Instead say the emissions were not included."

Reply - We have replaced "suppressed" with "turned off".

5. "Section 5.2, Page 32600, Lines 14 – 15: "Followed by a trend of decreasing with altitude" is awkward and should be reworded."

Reply - We have rewritten the sentence to "The ozone mixing ratio was lowest near the surface and increased with altitude below ~1.5km where it reached a maximum (94.7ppbv and 90ppbv in ozonesonde and aircraft measurements, respectively), and then decreased with altitude from ~1.5km to ~4km."

Reply to Referee #2's comments:

Thank Referee #2 for very helpful suggestions and comments.

1. "Only one comment to be made with a request for short discussion and references. Stratospheric intrusions are active during the springtime period of the study and can also add what looks like "background" ozone. This is acknowledged in the discussion of Fig 6 in the paper where in April 2005, especially, stratospheric contributions to ozone below 6 km can be very high. However, the possibility should be mentioned in the Introduction to the paper as well..."

Reply - Indeed. Following Referee #2 and M. Lin's suggestions, we have added the following text in the literature review: "Note, however, that stratospheric intrusions also maximize at mid-latitudes in spring. Ozonesonde measurements in western North America between 40°N and 55°N in April-May 2006 showed that stratospheric influences can be comparable to impacts of Asian transport (Doughty et al., 2011; Moody et al., 2012). Studies of the sources and variability of LT O₃ over China have also emphasized the roles of biomass burning emissions (Liu et al., 1999; Fu et al., 2007; Lin et al., 2009), biogenic emissions (Fu et al., 2007), and the impact of the Asian monsoon system (Liu et al., 2002; Lin et al., 2009)." Relevant references have also been added.

2. "Fix spelling errors. Page 6 – line 10 – ENSCI-ECC not ENSC-ECC Page 29 Suarez in the Rienecker ref."

Reply - Fixed.

Revised text with track changes

(next page)

Origin of springtime ozone enhancements in the lower troposphere over Beijing: In situ measurements and model analysis

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Abstract. Ozone (O_3) concentrations in the lower troposphere (LT) over Beijing have significantly increased over the past two decades as a result of rapid industrialization in China, with important implications for regional air quality and photochemistry of the background troposphere. We characterize the vertical distribution of lower-tropospheric (0-6km) O_3 over Beijing using observations from 16 ozonesonde soundings made during a field campaign in April-May 2005 and MOZAIC (Measurement of Ozone and Water Vapor by Airbus In-Service Aircraft) aircraft measurements over 13 days in the same period. We focus on the origin of LT O_3 enhancements observed over Beijing, particularly in May. We use a global 3-D chemistry and transport model (GEOS-Chem CTM) driven by assimilated meteorological fields to examine the transport pathways for O_3 pollution, and quantify the sources contributing to O_3 and its enhancements in the springtime LT over Beijing. Output from the Global Modeling Initiative (GMI) CTM is also used. High O_3 concentrations (up to 94.7 ppbv) were frequently observed at the altitude of ~1.5-2km. The CTMs captured the timing of the occurrences but significantly underestimated their magnitude. GEOS-Chem simulations and a case study showed that O_3 produced in the Asian troposphere (especially from Asian anthropogenic pollution) made major contributions to the observed O_3 enhancements. Contributions from anthropogenic pollution in the European and North American troposphere were reduced during these events, in contrast with days without O_3 enhancements, when contributions from Europe and North America were substantial. The O_3 enhancements typically occurred under southerly wind and warmer conditions. It is suggested that an earlier onset of the Asian summer monsoon would cause more O_3 enhancement events in the lower troposphere over the North China Plain in late spring and early summer.

1 Introduction

Tropospheric ozone (O_3) is an effective greenhouse gas, especially in the upper troposphere (UT) (Lacis et al., 1990). It is also the primary source of hydroxyl radicals (OH) that determine the oxidizing capacity of the atmosphere (Thompson, 1992). As an air pollutant near the surface, it has a detrimental effect on both vegetation and human health. Tropospheric O_3 has two sources - photochemical oxidation of hydrocarbons and carbon monoxide (CO) by OH radicals in the presence of oxides of nitrogen ($NO_x = NO + NO_2$), and downward transport from the stratosphere. Its precursors (NO_x , hydrocarbons, CO) are generated by fossil fuel combustion, industrial processes, biomass burning, vegetation, microbial activity in soils, and lightning.

Ozone concentrations have significantly increased in the lower troposphere (LT) across China (Chan et al., 2003; Ding et al., 2008; Wang et al., 2009) over the past decades as a result of increasing anthropogenic precursor emissions (Richter et al., 2005). Elevated O_3 levels not only lead to degradation of local and regional air quality (Wang et al., 2006; Wang et al. 2008; Lin et al., 2008), but also have significant implications for chemical environment and air quality in downwind regions (e.g., Hudman et al., 2004; [Lin et al., 2012, 2014](#)). Via deep convection or strong warm conveyor belts, Asian pollutants can be lifted up into the upper troposphere and transported to the North Pacific and North America (Jaffe et al., 1999; Liu et al., [2002](#), 2003; Liang et al., 2004; Cooper et al., 2010). This transpacific transport is most efficient in spring when cold fronts frequently occur and strong westerlies prevail in the upper troposphere over East Asia (Liu et al., 2003; Liang et al., 2004). Note, however, that stratospheric intrusions also maximize at mid-latitudes in spring. Ozone measurements in western North America between 40°N and 55°N in April-May 2006 showed that stratospheric influences can be comparable to impacts of Asian transport (Doughty et al., 2011; Moody et al., 2012). Studies of the sources and variability of LT O_3 over China have also emphasized the roles of biomass burning emissions (Liu et al., 1999; Fu et al., 2007; Lin et al., 2009), biogenic emissions (Fu et al., 2007), and the impact of the Asian monsoon system (Liu et al., 2002; Lin et al., 2009).

~~With a population of over 19 million and~~ Located in northern China ~~and with a population of over 19 million~~, Beijing is one of the world's largest cities and facing a severe problem of O₃ pollution. Ozone measurements over Beijing, derived from MOZAIC (Measurement of Ozone and Water Vapor by Airbus In-Service Aircraft) aircraft data from 1995 to 2005, showed that O₃ in the LT has increased by 2% annually and ~4% during May-July in contrast to flat or negative trends of other mega-cities at a similar latitude (Tokyo, New York, and Paris), and also exhibited higher daytime O₃ concentrations than these cities (Ding et al., 2008). Wang et al. (2012) reported a summertime increase rate of 3.4% yr⁻¹ in the lower-tropospheric (0-3km) partial O₃ column over Beijing based on ozonesonde measurements from 2002 to 2012. In addition to the long-term positive trend, O₃ over Beijing often experiences high-concentration episodes at ground level. Wang et al. (2006) reported that in 13 out of 39 days of surface observations at an elevated site (280 meters asl) in northern Beijing during June-July 2005, ambient O₃ levels exceeded 120 ppbv and had a 1-hr maximum level of 286 ppbv. The IASI (Infrared Atmospheric Sounding Interferometer) observed O₃ concentrations in the LT (3km) over Beijing up to 10 ppbv higher than the values for the 40-50°N latitude band climatology, which is more representative of background O₃ (Dufour et al., 2010). Dufour et al. reported that the lower-tropospheric partial (0-6km) column O₃ in Beijing shows a sharp increase in late spring with a maximum in May.

High ground-level O₃ in Beijing is caused not only by local anthropogenic emissions but also by regional and long-range transport. As reported by Streets et al. (2007), 35-60% of O₃ during high-O₃ episodes at Beijing Olympic Stadium originated from sources outside Beijing. Back-trajectory analysis by Ding et al. (2008) indicated that elevated O₃ levels in the boundary layer over Beijing during May-July were mostly related to emissions from the North China Plain. However, past investigations of the sources and transport of lower-tropospheric O₃ in Beijing have mostly involved categorizing wind directions or calculating back trajectories, techniques that do not allow its various sources to be quantified. In addition, previous studies were mainly based on surface measurements, which are not adequate for a full understanding of processes that control the vertical distribution and

variability of lower-tropospheric O₃ in Beijing.

To characterize the distribution and variability and quantify the sources of springtime lower-tropospheric O₃ in Beijing, we analyze O₃ vertical profiles measured during an ozonesonde sounding campaign, as well as aircraft measurements made by MOZAIC program. We show that O₃ enhancements in the LT over Beijing were frequently observed by ozonesondes during April-May 2005. Such O₃ enhancements during some of those days were also captured by the MOZAIC aircraft. We apply the GEOS-Chem chemistry and transport model (CTM), driven by assimilated winds, to quantify the sources contributing to these O₃ enhancements over Beijing and examine the corresponding pollution transport pathways in East China. By tagging O₃ originating in different source regions and conducting sensitivity simulations, we show that these O₃ enhancements were mainly due to Asian anthropogenic pollution, while the impact of the European and North American emissions were significantly smaller. We find that the high O₃ episodes occurred mostly under southerly wind conditions. We also evaluate the Global Modeling Initiative (GMI) CTM simulations with observations, and show that current global models significantly underestimated the magnitude of these O₃ enhancements.

This paper is structured as follows. The ozonesonde and aircraft measurements, as well as GEOS-Chem and GMI CTM are briefly described in Section 2. We describe the characteristics of the distribution and variability of springtime lower-tropospheric O₃ in Beijing from ozonesonde observations and MOZAIC aircraft measurements in Section 3. The model performance in reproducing the observed characteristics is examined in Section 4. The model analysis and case study of various sources of lower-tropospheric O₃ enhancements are presented in Section 5, followed by summary and conclusions in Section 6.

2 Data and methods

2.1 Ozonesonde and aircraft measurements

We use two sets of in-situ O₃ vertical profiles over Beijing obtained during April-May

2005: One from the Transport of Air Pollutants and Tropospheric Ozone over China (TAPTO-China) (Chan et al., 2007; Zhang et al., 2012) ozonesonde campaign, and the other from the MOZAIC aircraft program (Marenco et al., 1998). The concurrent availability of these two data sets provides an excellent opportunity for cross-validation.

TAPTO-China was an intensive ozonesonde sounding campaign over China, conducted at five locations across southern China in the spring of 2004 (Zhang et al., 2012) and four locations across northern China in the spring of 2005. The main objective of TAPTO-China was to investigate the mechanisms controlling the spatio-temporal distribution, variability, and sources of springtime tropospheric O₃ over China and its surrounding regions. Figure 1 shows the locations of the four ozonesonde stations in northern China during the second phase of TAPTO-China (April-May 2005): Xining (36.43°N, 101.45°E), Beijing (39.80°N, 116.18°E), Longfengshan (44.44°N, 127.36°E), and Aletai (47.73°N, 88.08°E). In this [study](#) [westudy](#), we use the ozonesonde sounding data obtained at the Beijing station (34 meters asl), which is located in the suburban Daxin district of southeastern Beijing. Sixteen ENSCI-ECC ozonesonde (with Vaisala RS 80 radiosonde) soundings in total were performed at this station during April 11 – May 15, 2005, with one sounding every 2-3 days on average. These sondes were launched at 13:30 local time, and the air was sampled every 15s. Pollution at the station [wa](#)is expected to be heavier than the average in the city because it is located [at](#)-downwind of Beijing. All raw ozonesonde data were averaged into 100m bins.

The MOZAIC program was initiated in 1993 to collect experimental data of O₃ and water vapor obtained by five long-range commercial airlines flying all over the world. Its goal was to provide a large database for studies of atmospheric chemical and physical processes in order to improve chemistry and transport models (Marenco et al., 1998). Dual-beam UV absorption analyzers (Model 49-103 from Thermo Environment Instruments) were installed onboard the aircraft to measure O₃, and were calibrated in the laboratory every 6-12 months with an overall precision of +/- (2ppb + 2%) (Thouret et al., 1998; Thouret et al., 2006). Tropospheric O₃ measurements were routinely conducted during the ascents and descents of flights nearby 50 cities frequented by MOZAIC operation (Marenco et al., 1998), including

Beijing. More information about the MOZAIC program can be found on its website (<http://www.iagos.fr/web/rubrique362.htm>). The Beijing Capital International Airport is located about 25 km southeast of the urban Beijing area, and is surrounded by the mountains to the west, north, and northeast (Ding et al., 2008). MOZAIC obtained vertical O₃ profiles in Beijing from March 1995 through March 2006. For April-May 2005, 13 profiles are available. The raw data were sampled every 4s (~30m) vertically and averaged into 150m bins.

2.2 CTM simulations

GEOS-Chem is a global 3-D model of tropospheric chemistry driven by assimilated meteorological observations from the Goddard Earth Observing System (GEOS) of the NASA Global Modeling and Assimilation Office (GMAO). The use of assimilated meteorological data to drive the model makes it an ideal tool for explaining the factors governing observed constituent distributions for a specific year. The description and evaluation of GEOS-Chem as applied to tropospheric O₃-NO_x-hydrocarbon chemistry was first presented by Bey et al. (2001b), and a description of the coupled oxidant-aerosol simulation was given by Park et al. (2004). The reader is referred to the Appendix for a brief description of other aspects of the model, including the emissions used. Here, we use GEOS-Chem version v9-01-02 (<http://www.geos-chem.org>) driven by two generations of GEOS assimilated meteorological fields, GEOS-4 (Bloom et al., 2005) and GEOS-5 (Rienecker et al., 2008) with a degraded horizontal resolution (2°×2.5°). The two sets of meteorological input data allow for examination of the sensitivity of model results to uncertainties in our characterization of the April/May 2005 meteorology. While GEOS-Chem is the major modeling analysis tool we use, we also use outputs from simulations made with GMI CTM (<http://gmi.gsfc.nasa.gov>) (e.g., Strahan et al., 2007; Duncan et al., 2007; Considine et al., 2008; Allen et al., 2010) to take into account the uncertainty in model simulations. GMI CTM combines both tropospheric and stratospheric chemical mechanisms. GMI simulations are driven by the GEOS-4 and MERRA (i.e., GEOS-5.2.0) meteorological data sets (2°×2.5°).

We use GEOS-Chem to investigate the contributions to the LT O₃ in Beijing from six source regions: troposphere of Asia, Africa, Europe, North America, the stratosphere, and the rest of the world. Figure 2 shows the tropospheric O₃ source regions defined in the model for tagged O₃ simulations. Ozone produced in each of these source regions is transported separately in the model, and removed by chemical loss and dry deposition at the same frequencies as those for total O₃. The sum of all tagged O₃ tracers is equivalent to the O₃ concentration from the standard full-chemistry simulation (Wang et al., 1998). This approach was previously applied to a number of investigations (e.g., Liu et al., 2002; Liu et al., 2009; Zhang et al., 2012). However, it is important to note that tagged O₃ by source regions does not represent the sensitivity of tropospheric O₃ to emissions in those regions. This is because O₃ precursors themselves can be transported out of their source regions and can therefore contribute to O₃ production elsewhere.

We therefore also conduct sensitivity simulations to examine the effect of various emission types on tropospheric O₃ over Beijing. In these simulations, emissions from Asian fossil fuel, biomass burning, European fossil fuel, North American fossil fuel, and lightning NO are individually shut off to quantify their contributions to O₃. Simulations were conducted from August 2004 to May 2005 with the first eight months being used for initialization.

Spring in East Asia is a meteorological transition season when the winter monsoon retreats and summer monsoon gradually marches northward. Figure 3 shows the average wind vectors in the LT (~870hPa) during April-May 2005. Superimposed colors indicate the model average O₃ mixing ratios. The circulation patterns and O₃ distributions in the two simulations with GEOS-4 and GEOS-5 show similar features. In East China, the northwesterly and southwesterly winds converge around 30°-40°N (North China Plain), flanked by the western Pacific subtropical high to the east. In this convergence zone, surface pollutants can be readily lifted out of the boundary layer and transported to the western Pacific by westerly winds in the free troposphere (Bey et al., 2001a; Liu et al., 2003). Beijing is located in the northern part of the North China Plain, where northwesterly winds prevail

near the surface in spring. However, as the Siberian High weakens and the East Asia summer monsoon develops towards late spring, incursions of warmer tropical air from the south become more frequent and vigorous, especially in May (Ding and Chan, 2005), putting Beijing under the influence of southerly air masses.

3 Lower-tropospheric O₃ in Beijing as observed by ozonesonde and aircraft

In this section, we examine the distribution and variability of springtime lower-tropospheric O₃ in Beijing using ozonesonde and MOZAIC aircraft measurements for April-May 2005. ~~Both types of measurements were made by both platforms on four of all~~ days: May 1, ~~May 3, May 11, and May 15~~. The vertical distributions of O₃ in the LT in the two data sets are consistent on each of these four days. This is illustrated in Figure 4 (panel a) with the four-day average profile. The consistency indicates that both ozonesonde and MOZAIC measurements well captured tropospheric O₃ concentrations over Beijing.

Figure 4 (panels b and c) shows the mean vertical distribution of O₃ below 6km as calculated from the April/May ozonesonde and aircraft data in comparison with model simulations. Model results will be discussed later in Section 4. The average O₃ distribution observed in the LT over Beijing ranged from 44 to 67 ppbv with a minimum near the surface and an enhancement at ~1.5km. Ozone concentrations remained fairly constant (~65ppbv) above ~1.5km. This LT enhancement in the vertical O₃ distribution over Beijing is consistent with the MOZAIC climatology (1995-2005) where O₃ concentrations peak below 2km in May-July, but the peak in the latter was located at a somewhat lower altitude (~1.0km) (Ding et al., 2008).

The low average O₃ concentration observed near the surface (below 1km) over Beijing (Fig. 4bc) reflects the frequent low O₃ concentrations below 1km seen in the individual sonde or aircraft profiles (Fig. 5). The minimum LT O₃ concentration seen in the ozonesonde observations was a value of 0.2 ppbv at 0.3km on May 13; on this day, O₃ mixing ratios were lower than 2ppbv up to 0.6km. Low-O₃ episodes were also observed near the surface on Apr

17, April 23, April 29, and May 9. These low O₃ mixing ratios are likely due to the chemical titration by high NO_x concentrations, a common characteristic in urban areas (e.g., Chan, et al., 1998). Backward trajectory calculations (not shown) suggest that cleaner air masses originating from the east may also contribute to the low O₃ concentrations seen here.

The O₃ concentrations (~65 ppbv) at ~1.5 km in the average profile reflects O₃ enhancements (> 70 ppbv) frequently observed in individual profiles during April-May, 2005. Enhanced O₃ mixing ratios were observed below 4km in 15 out of total 25 days with observations: April 7, April 13, April 21, April 23, April 27, April 29, May 1, May 3, May 11, May 12, May 13, May 15, May 18, May 24, and May 29 (Fig. 5). Of these 15 days, 11 days (i.e., all except April 7, April 21, May 1, and May 13) saw enhanced O₃ mixing ratios at ~1.5km over Beijing. Relatively large O₃ enhancements at this altitude were mostly observed in late April and May, such as April 29, May 3, May 11, May 12, May 15, May 24, and May 29. The maximum O₃ concentration observed in the LT was 95 ppbv at ~1.4km on May 3 during the ozonesonde sounding campaign, and 131 ppbv at ~2.3km on May 12 as observed by MOZAIC aircraft. Ozone enhancements on May 3, May 11, and May 15 were observed simultaneously by ozonesonde and MOZAIC aircraft.

These aforementioned springtime O₃ enhancements are likely to be due to regional photochemical pollution. Recent studies indicate that transport of pollutants from surrounding regions have an important influence on air quality in Beijing during spring (Zhang et al. 2006; An et al., 2007). A regional model simulation of an air pollution episode in Beijing during April 3-7, 2005 by An et al. (2007) showed that non-Beijing sources contributed about 39% to PM_{2.5} and 60% to PM₁₀ concentrations in Beijing. Lin et al. (2008) showed that the North China Plain contributed 19.2 ppbv to the surface O₃ at a rural site north of Beijing (Shangdianzi, one of the regional Global Atmosphere Watch stations in China) in spring. Nevertheless, intercontinental transport may also contribute to these O₃ enhancements. We will address this issue in Section 5.

These springtime O₃ enhancements over Beijing are also likely related to favorable meteorological conditions. LT temperature inversions were observed in sonde profiles over

Beijing on April 11, April 23, April 27, April 29, May 1, May 3, May 9, May 13, and May 15 (not shown). Particularly, strong inversions were found at ~1.5km on April 29 and May 3, when relatively large O₃ enhancements were seen at the same altitude. When such inversions are present, vertical mixing is suppressed, trapping O₃ and its precursors below the inversion. This is similar to the elevated levels of O₃ previously detected above the base of temperature inversions in and nearby the similarly polluted Los Angeles Basin (Lea, 1968; Edinger, 1973; Blumenthal et al., 1978).

The LT over Beijing is near the boundary between regions of southwesterly (southerly) and northwesterly winds during spring (Fig. 3). The transition of winter-to-summer monsoon during this period has an important impact on the transport of O₃ in the LT. Trajectory classifications over Beijing during May-July, as shown by Ding et al. (2008), revealed that the O₃ concentrations in the air masses from the south are 10-15ppbv higher than those from the north below 2km, and the difference was largest at 1km. By comparing surface O₃ concentrations observed at Shangdianzi under southwest (SW) and northeast (NE) wind conditions, Lin et al. (2008) showed that the average O₃ concentrations corresponding to the SW wind directions were higher than those of NE wind directions in spring, and the difference was over >20ppbv in May. We will illustrate such influences of meteorological conditions on the LT O₃ with a case study to be presented in Section 5.

4 Model simulations of lower-tropospheric O₃ over Beijing

In this section, we present model simulations of LT O₃ over Beijing in comparison with in situ (ozonesonde and aircraft) measurements. Because the elevation of Beijing in the model is 290 meters asl, which is higher than the actual elevation (160 meters asl), there is a small gap below 290 meters where O₃ is not simulated. Figure 4bc shows the model-observation comparison of the average vertical distribution of O₃ in the LT during April-May 2005. Results from both GEOS-Chem (driven by the GEOS-4 and GEOS-5 meteorological fields) and GMI (driven by the GEOS-4 and MERRA meteorological fields) models are shown.

Figure 5 shows the time-height cross-sections of the LT O₃ mixing ratios (ppbv), as observed by ozonesonde and aircraft and simulated by GEOS-Chem during this period.

The simulated mean vertical distributions of O₃ are similar to the observations (Fig. 4bc). The GEOS-Chem/GEOS-4 model agrees with the observations near 3-4.5km, but underestimates the observations by 2 ppbv between ~1-2.5km and overestimates O₃ by 1.7-7.7ppbv below 1km and by 1.3-6.7ppbv above 4.5km, with a large bias of ~8ppbv near the surface (Fig. 4b). The GEOS-Chem/GEOS-5 simulated O₃ is in good agreement with observation above 2.5km but ~~underestimates the observations at ~1-1.5km too low~~ by up to 6ppbv at ~ 1-1.5km compared to observations (Fig. 4c). Both simulations showed negative biases around 1.5km, with the GEOS-Chem/GEOS-4 values in slightly better agreement. Both simulations fail to reproduce the magnitude of the daily LT O₃ enhancements over Beijing, as discussed further below.

The GMI model simulations driven by GEOS-4 and MERRA meteorological fields shown in Figure 4(bc) better reproduced than GEOS-Chem the observed values at ~0.5km. Above ~2.5km, the GMI and GEOS-Chem simulations driven by the GEOS-4 meteorology are consistent. Near 1.5km, however, GMI/GEOS-4 has a larger negative bias than GEOS-Chem (Fig. 4b). The GMI/MERRA model consistently underestimates O₃ observations below 4.5km, with the largest bias near 1.5km (~10ppbv) and larger biases than those of GEOS-Chem/GEOS-5 (Fig. 4c).

Both GEOS-Chem/GEOS-4 and GEOS-Chem/GEOS-5 simulations capture the large-scale temporal (day-to-day) variability in the vertical distribution of O₃ seen in the ozonesonde and aircraft measurements (Fig. 5). They well simulate the O₃ enhancement event at ~1.5km on May 3, which is the largest observed by both ozonesonde and MOZAIC aircraft at this altitude during this period. The timing of O₃ enhancements, especially those observed in late April and May, is reasonably captured. However, the model is missing the April 23rd enhancement and the April 27th event is shifted to April 29th in the model. Overall, the model underestimates the magnitude of those O₃ enhancements. Also, both simulations fail to reproduce the observed low-O₃ episodes near the surface, where the O₃ concentrations

are largely overestimated. Model simulations at higher horizontal resolutions that better resolve the chemical titration of O₃ by excessive NO_x may help address this discrepancy.

The distribution and variability of model relative humidity (RH) are generally similar to those observed by ozonesondes (not shown), suggesting that convective transport and large-scale ascending and descending motions in the study region are reasonably represented in the GEOS-4 and GEOS-5 meteorology. GMI/GEOS-4 and GMI/MERRA simulations also captured the observed large-scale variability in O₃ and timing of O₃ enhancements (not shown).

5 Sources of springtime O₃ enhancements in the lower troposphere over Beijing

In this section we quantify the relative contributions to the LT O₃ over Beijing of different source types and source regions in the GEOS-Chem model. We focus on model simulations driven by GEOS-4 but also discuss GEOS-Chem/GEOS-5 results where necessary. The relative contributions allow us to better understand the factors and processes contributing to the observed O₃ enhancements and distributions in the LT, and therefore may provide insights into the model limitations in the representation of chemical, physical, and dynamical processes. Figure 6 shows the major sources of the LT O₃ over Beijing during April-May 2005 as a function of date and altitude, as simulated by GEOS-Chem/GEOS-4. The plots show model results for tagged O₃ tracers transported from the stratosphere and those produced in the Asian, African, European, and North American troposphere. Figure 7 presents the changes (decreases) in the LT O₃ concentrations over Beijing when Asian, European, North American fossil fuel emissions, global biomass burning emissions, or lightning NO_x emissions were suppressed/turned off, respectively, in GEOS-Chem/GEOS-4 and GEOS-Chem/GEOS-5, relative to their standard simulations. Shown in Figure 8 are the time series of temperature, total tagged O₃, and tagged O₃ tracers concentrations in the LT (~878hPa or 840-915hPa in the model) over Beijing, as simulated by GEOS-Chem/GEOS-4.

These figures provide a context for most of the discussions below.

5.1 Contributions to O₃ in the LT over Beijing of different source regions and emission types

Ozone produced within Asia made a major contribution to the LT O₃ over Beijing during April-May 2005. The Asian troposphere contributed ~10-46.6ppbv (~17-66%) in early-mid April and ~9-81ppbv (~15-88%) in late April and May (Fig. 6, top panels). The contribution was significantly higher in May than in April. This is consistent with the earlier result that relatively large enhancements of O₃, which were observed by ozonesonde and aircraft and captured in model simulations, mostly occurred in late April and May. On the dates of these O₃ enhancement events (e.g., April 29, May 3, May 13, and May 15), LT O₃ was predominantly produced within Asia (~60-79.4ppbv, or ~81%-88%). A modeling study by Sudo and Akimoto (2007) also suggested that LT O₃ in Japan and coastal China regions during TRACE-P (spring 2001) was mainly produced within Asia and mostly in the planetary boundary layer.

The aforementioned relatively large enhancements of O₃ were largely due to Asian fossil fuel emissions. When Asian fossil fuel emissions were ~~suppressed~~turned off, the LT O₃ concentrations over Beijing decreased by ~3.1-21.3ppbv in the GEOS-Chem/GEOS-4 simulation and by ~3.0-25.3ppbv in the GEOS-Chem/GEOS-5 simulation in early-mid April, and decreased by ~2.2-42.4ppbv and ~3.0-37.8ppbv, respectively, in late April and May (Fig. 7, top panels). The impact of Asian fossil fuel emissions was stronger in late April and May, especially during those days with large enhancements of O₃. Without Asian fossil fuel emissions, the LT O₃ reduced by ~30-40 ppbv on April 29, May 3, May 13, and May 15. To understand the influence of fossil fuel emissions (or O₃ produced) in the sub-regions within Asia on the LT O₃ over Beijing, additional sensitivity or tagged O₃ simulations with refined emission (or O₃ source) regions would be required. Nevertheless, the case study to be presented in the following section will provide insights into the effect of regional pollution

transport in this regard.

Stratospheric contributions to the LT O₃ over Beijing during April-May 2005 were significant (Fig. 6). They were ~6.0-13.0ppbv (~8.5-20.6%) in April, and ~1.6-19.8ppbv (~2.1~23.8%) in May, and increased with altitude. On average, stratospheric contributions were larger in April (~7.6ppbv) than in May (~5.7ppbv). At the time of occurrences of the aforementioned O₃ enhancement events, stratospheric contributions were significantly reduced below ~3km (~2ppbv, ~2%). The relative magnitude of the contributions of O₃ produced within the African troposphere was lower than that of the stratospheric contributions (Fig. 6). The African troposphere contributed ~2.4-16.5ppbv (~3-28%) O₃ in April and ~0.9-10.6ppbv (~1-13%) O₃ in May.

The impact of biomass burning emissions on the LT O₃ over Beijing during this period was relatively small. When biomass burning emissions are suppressturned off in GEOS-Chem, O₃ concentrations decrease by ~1±0.5ppbv in April and by ~2±1ppbv in May, with a maximum decrease of ~6.7ppbv on April 7 (Fig. 7). When lightning NO_x emissions are suppressturned off, the LT O₃ concentrations fall by ~1.3-9.1 ppbv in the GEOS-Chem/GEOS-4 simulation and by ~1.5-14.5ppbv in the GEOS-Chem/GEOS-5 simulation in April; in May, they decrease by ~0.3-14.3ppbv and by ~0.5-21.3ppbv in May, respectively (Fig. 7). The larger impact of lightning NO_x emissions in May reflects the seasonal increase in continental deep convection, which results in more frequent lightning. Relatively small effects (~3±1ppbv) were seen at ~1.5km, with an effect of ~0.9±0.1ppbv O₃ at the time of large enhancements of O₃.

The European and North American source regions and their anthropogenic emissions have important impacts on LT O₃ over Beijing. Europe contributed ~2-16.7ppbv (~3-29.4%) of O₃ in April and ~2.5-20.8ppbv (~3-29.9%) in May (Fig. 6). The contributions to LT O₃ below 4km were larger in April. For these large enhancements of O₃, the contributions from Europe were significantly reduced. Similarly, North America contributed ~2.9-16.6 ppbv (~3.9-26.5%) in April and ~2.5-20.3 ppbv (3.2-26.1%) in May, with much smaller contributions during O₃ enhancement events (Fig. 6). When European fossil fuel emissions were suppressturned off,

the LT O₃ concentrations over Beijing decreased by ~1.3-10 ppbv in April and ~1-8 ppbv in May (Fig. 7). The decreases were ~1-6 ppbv in April and ~0.6-6 ppbv in May when North American fossil fuel emissions were ~~suppressed~~ turned off. Again, the impacts of European and North American fossil fuel emissions were minimal (<~1ppbv) during the events of large enhancements of O₃.

Figure 8 shows the time series of total O₃, tagged O₃, and temperature in the LT (~878 hPa, ~1.5km) over Beijing in GEOS-Chem/GEOS4. Occurrences of these O₃ enhancements were accompanied by significantly increased contributions of O₃ produced in the Asian troposphere and often associated with warm air masses. On the days with relatively large observed O₃ enhancements (April 29, May 3, May13, and May 15), the contributions of O₃ from the Asian troposphere were 62.1ppbv (77.4%), 76.0ppbv (87.0%), 79.4ppbv (86.4%), and 64.0ppbv (87.2%), respectively. By contrast, at the time of these events, the contributions from Europe and North America were largely reduced. Also, compared to the more prominent contributions from the Europe (~11.6ppbv) and North America (~11.6ppbv) in early April, they were relatively smaller in late April and May, at an average of ~8.2ppbv and ~8.0ppbv, respectively.

As Beijing is located at mid-latitudes in East Asia (40°N), air masses originating from Europe and North America arrived as a result of large-scale subsidence associated with the Siberian anticyclone. Previous studies have shown that European anthropogenic emissions contribute significantly to the Asian outflow in the LT at latitudes north of 35°N (Bey et al., 2001a; Liu et al., 2003). As the Siberian high gradually weakened in spring and its center shifted westward, air masses from these regions had a reduced influence. Meanwhile, the western Pacific subtropical high developed and its center marched northwestward. As a result, ~~The~~ the northward transport of warmer air masses from South China and along the Asian Pacific rim became more vigorous. An examination of the GEOS-4 and GEOS-5 meteorological fields shows that Beijing was mostly under the influence of southwesterly flow on the days with observed ozone enhancements. This is consistent with the stronger influence of Asian sources and more frequent occurrences of large O₃ enhancements in late

April and May.

5.2 Ozone enhancements in the lower troposphere: A case study

In order to better understand the transport pathways for O₃ pollution and the vertical extent of O₃ enhancements over Beijing, we examined the largest O₃ enhancement observed by both TAPTO-[China](#) ozonesonde and MOZAIC aircraft during April-May 2005. Figure 9 shows the vertical profiles of O₃ over Beijing from TAPTO ozonesonde and MOZAIC aircraft measurements in comparison with those simulated by GEOS-Chem/GEOS-4 and GMI/GEOS-4 on May 3, 2005. Also shown are observed RH, temperature, and O₃ sources as determined by GEOS-Chem tagged O₃ and sensitivity simulations.

Ozonesonde and aircraft measurements of O₃ (0-6km) agree within ~2% on average and show similar structures in their vertical distributions. The ozone mixing ratio was lowest near the surface and increased with altitude below ~1.5km where it reached a maximum (94.7ppbv and 90ppbv in ozonesonde and aircraft measurements, respectively), ~~followed by a trend of decreasing with altitude up and then decreased with altitude from ~1.5km~~ to ~4km. The rapid increase of O₃ concentrations from ~1km up to ~1.5km was accompanied by a temperature inversion observed by ozonesonde. This temperature inversion inhibited the vertical mixing and transport of the air, trapping the air mass containing high levels of O₃ (and its precursors) at this level. The presence of the inversion also suggests that the high O₃ at ~1.5km was not transported from the local boundary layer via vertical mixing. Rather, backward trajectory calculations using the NOAA HYSPLIT model (Draxler and Rolph, 2014; Rolph, 2014) show that the air mass was transported downward from a higher altitude (~3km) and stayed at about 1.4km for two days before arriving at the 1.4km level over Beijing (Fig. 10).

The relatively low O₃ below 1km was likely due to chemical titration by local fresh NO_x emissions. The 5-day backward trajectory arriving at 150m over Beijing indicated that the air mass stayed near the surface during the days prior to its arrival (Fig. 10), providing optimal conditions for chemical titration.

The GEOS-Chem/GEOS-4 simulation shows an enhancement of O₃ at ~1.5km on May 3rd, but significantly underestimates its magnitude by ~10-15ppbv (Fig. 9). This is partly attributed to the inability of a large-scale model to reproduce the local temperature inversion (not shown). The simulation reasonably reproduced the observations above 2.5km, but overestimated the observations (by ~5-10ppbv) below 1km, presumably because chemical titration was not well represented and there is too much vertical mixing due to lack of a temperature inversion in the model. The GMI/GEOS-4 simulation was better reproducing the relatively low O₃ concentrations near the surface. However, it fails to capture the O₃ enhancements at ~1.5km. The LT O₃ concentrations simulated by GMI/GEOS-4 are much lower than the observations, with the largest discrepancy (~26.9ppbv) at ~1.5km.

Model simulations suggest that this LT O₃ enhancement over Beijing was largely due to Asian fossil fuel emissions. Without Asian fossil fuel emissions in the GEOS-Chem/GEOS4 model, the simulated O₃ concentration in the LT decrease by ~30% (Fig. 9). The impact of the Asian fossil fuel emissions is largest (~30ppbv, 40%) at ~1.5km where LT O₃ concentrations reached a maximum. In addition, tagged O₃ simulations showed that 79.6% of O₃ at ~1.5km originated from the Asian troposphere (not shown). When lightning NO_x emissions were suppressed, the decreases of the LT O₃ over Beijing increased with altitude, with the smallest decreases (~1.2ppbv, ~1.7%) below ~1.5km. Similarly, the contributions of stratospheric O₃ increased with altitude, with a small contribution (~2.9ppbv, ~4%) at ~1.5km where the largest enhancement of O₃ occurred. Therefore, this LT O₃ enhancement was mainly due to the regional anthropogenic pollution within Asia.

The GEOS4 assimilated meteorological data driving the GEOS-Chem model indicate that strong southwesterly winds prevailed over Beijing in the LT on May 2-3 (Fig. 11, Fig. 12). Meanwhile, a low-pressure center in the LT at (~43°-53°N, 120°-140°E) put the area north of Beijing under the influence of northwesterly winds. The southwesterly and northwesterly winds met just north of Beijing at around 43°N, forming a convergence zone. As part of the western Pacific subtropical high ~~that was~~ developing around 15°-30°N, strong southwesterly winds over southern and eastern China could have readily swept the pollutants in its pathway

into Beijing. Indeed, an area of high CO (>200 ppbv) was located to the southwest of the North China Plain on May 2-3 at 878hPa. The air mass stayed in this high CO area for a couple of days before arriving at 1.4km over Beijing (Fig. 10), where the O₃ enhancement was observed by both sonde and aircraft, as well as simulated by the model. By contrast, the air mass arriving at 2.4km came straight from the west where air was cleaner and thus had significantly lower O₃. The high CO area was largest near the surface and became smaller with altitude. High CO concentrations were seen in the model simulations up to ~ 797 hPa. No high-CO plumes were found at ~ 704 hPa over East Asia on May 2-3 in the model (not shown). By May 3, Beijing was at the center of the high O₃ area (>72 ppbv) in the LT (797hPa and 878hPa, Fig. 12) on May 3. The O₃ concentrations were also relatively lower ($\sim 68 \pm 4$ ppbv) near the surface (942hPa) and at a higher altitude (704hPa).

6 Summary and conclusions

We have characterized the vertical distribution of lower-tropospheric O₃ over Beijing using observations from 16 ozonesonde soundings made during the TAPTO-[China](#) field campaign in April-May 2005, as well as MOZAIC aircraft ascending and descending profiles over 13 days in this period. A particular focus was placed on the origin of O₃ enhancements in the lower troposphere (LT, 0-6km). We used a global 3-D chemistry and transport model (GEOS-Chem CTM) driven by assimilated meteorological fields from the NASA Goddard Earth Observing System (GEOS-4 and GEOS-5) to interpret these characteristics and quantify the sources contributing to O₃ and its enhancements in the springtime LT over Beijing. We also used the output of the Global Modeling Initiative (GMI) CTM for comparison with O₃ observations.

The average observed profile of O₃ mixing ratio in the LT over Beijing during April-May, 2005 ranged from ~ 44 to 67ppbv with an enhancement at the altitude of ~ 1.5 km and a relatively low abundance near the surface. The relatively high average O₃ concentrations at ~ 1.5 km were due to frequently observed elevated O₃ levels (up to 94.7 ppbv in soundings

and 130.3ppbv in MOZAIC aircraft measurements). Such enhancements were observed at this altitude in 15 out of 25 days with observations during this period. Ozone enhancements were more frequently observed by ozonesonde in late April and May relative to early and mid- April. This relates to northward transport of pollution in East China to the LT over Beijing due to weakening of the Siberian high and the onset of the Asian summer monsoon. It also suggests that an earlier onset of the Asian summer monsoon would cause more O₃ enhancement events in the LT over the North China Plain in late spring and early summer.

The GEOS-Chem model driven by GEOS-4 and GEOS-5 assimilated meteorological fields adequately reproduced the average vertical distribution of LT O₃ and captured the timing of the observed O₃ enhancement events over Beijing during April-May 2005, but both simulations underestimated the magnitude of these enhancements. On the other hand, GEOS-Chem overestimated the observed low O₃ concentrations near the surface. The GMI model driven by GEOS-4 and MERRA better resolved the average O₃ concentrations near the surface, but was less successful in reproducing the enhancement of average O₃ concentrations at ~1.5km. Nevertheless, GMI did capture the timing of the major O₃ enhancement events observed. Both GEOS-Chem and GMI were successful in reproducing temporal fluctuations of observed O₃ because they are driven by assimilated meteorological fields. The fact that they did not capture the surface low or the 1.5km peak likely resulted from a combination of inability to represent the chemical regime at the surface (i.e., NO_x-driven titration) due to low spatial resolution, and overly vigorous vertical transport (i.e., lack of a temperature inversion at the altitude of ~1.5km).

We investigated the sources contributing to the LT O₃ enhancements observed over Beijing during April-May 2005 by tagging O₃ produced in different source regions in the GEOS-Chem/GEOS-4 model and by conducting model sensitivity calculations. We found that these O₃ enhancements were predominantly attributed to O₃ produced within Asia, especially for relatively large enhancements (~81~88%). For the large enhancements, the O₃ concentrations at ~1.5km were reduced by ~30-40ppbv when Asian fossil fuel emissions were suppressed within the model. Contributions of O₃ from outside Asia were relatively

small during these O₃ events. However, long-range transport of O₃ (and its precursors) from Europe and North America has important impacts on LT O₃ over Beijing during non-event periods, especially in April. The two alternating regimes of transport reflect the springtime transition from winter to summer monsoon in East Asia.

A case study of the O₃ enhancements at ~1.5km observed on May 3, 2005 showed that the event was largely attributed to Asian fossil fuel emissions (34%). Backward trajectory calculations suggested that the air mass stayed in the LT to the southwest of Beijing for a couple of days before arriving at ~1.4km over Beijing. Elevated CO levels were found in the model results on May 2nd and 3rd in the area where the air mass remained before reaching Beijing. The model simulations showed that Beijing was at the center of the high O₃ area in the LT (~878hPa to 797hPa) by May 3, with prevailing strong southwesterly winds that swept O₃ and its precursors downwind to Beijing.

We find that the ozonesonde and aircraft observations of springtime O₃ in the LT over Beijing are qualitatively consistent with the current understanding of tropospheric chemistry and transport as represented by the GEOS-Chem model. Using the same model driven by different meteorological fields (GEOS-4 and GEOS-5) does not alter our conclusion. However, current global models (e.g., GEOS-Chem and GMI) have difficulty in quantitatively reproducing the observed enhancements and depletions of the LT O₃ over Beijing. Model simulations at higher resolutions would better resolve regional meteorology and transport with respect to the topography in the North China Plain, and chemical transformation such as the titration of O₃ by NO. Models could also be further evaluated by increasing satellite observations as well as multi-year ozonesonde (Wang et al., 2012) and aircraft measurements (Ding et al., 2008). Such efforts would improve our quantitative understanding of the origin of tropospheric O₃ and elevated O₃ events over the North China Plain, allowing a better projection of regional O₃ pollution and its global impact.

Appendix A: GEOS-Chem CTM

An earlier version of the GEOS-Chem model that we applied to the analysis of ozonesonde O₃ observations was described in the appendix of Zhang et al. (2012). A similar description is included here for completeness, but for version v9-01-02 and with a different configuration. We drive the GEOS-Chem model with the GEOS-4 and GEOS-5 assimilated meteorological observations from the Goddard Earth Observing System (GEOS) of the NASA Global Modeling and Assimilation Office (GMAO). The original description and evaluation of GEOS-Chem as applied to tropospheric O₃-NO_x-VOC chemistry was presented by Bey et al. (2001a) and a description of the coupled oxidant-aerosol simulation was given by Park et al. (2004). A number of previous studies have applied the model to tropospheric O₃ in various regions of the world, including the Asian and western Pacific (e.g., Bey et al., 2001b; Liu et al., 2002), the Middle East (e.g., Li et al., 2001; Liu et al., 2009), the United States (e.g., Fiore et al., 2002; Zhang et al., 2008; Hudman et al., 2009), the Atlantic (e.g., Li et al., 2002), and the tropics (e.g., Martin et al., 2002; Nassar et al., 2009; Zhang et al., 2011).

The temporal resolution of the GEOS-4 and GEOS-5 meteorological data is 6 hours (3 hours for surface variables and mixing depth). GEOS-4 has a native resolution of 1° latitude by 1.25° longitude with 55 vertical levels between surface and 0.01hPa and 8 vertical levels between 0-6km. GEOS-5 has a higher native resolution of 0.5° latitude by 0.667° longitude with 72 levels in the vertical and 23 vertical levels between 0-6km. For computational expediency, horizontal resolutions are degraded to 2° latitude by 2.5° longitude for input to GEOS-Chem. In addition, vertical levels above 50hPa (GEOS-4) or 80hPa (GEOS-5) are merged to retain 30 (GEOS-4) or 47 (GEOS-5) vertical levels in total. A major difference between GEOS-4 and GEOS-5 is the parameterization scheme for convection. GEOS-4 uses the parameterization scheme of Zhang and McFarlane (1995) and Hack (1994) for deep convection and shallow convection, respectively. Convection in GEOS-5 is based on the relaxed Arakawa-Schubert parameterization scheme (Moorthi and Suarez, 1992). Previous model simulations of ²²²Rn (Zhang et al., 2011) and CO (Liu et al., 2010) indicate that tropical deep convection is significantly deeper in GEOS-4 than in GEOS-5. GEOS-Chem assumes rapid vertical mixing within the mixing layers diagnosed by GEOS.

The GEOS-Chem model includes 87 chemical species and transports 43 chemical tracers to describe tropospheric O₃-NO_x-VOCs-aerosol chemistry. Anthropogenic emissions are based on the Global Emission Inventory Activity (GEIA) (Benkovitz et al., 1996), overwritten with NO_x, CO and SO_x from the Emission Database for Global Atmospheric Research (EDGAR) inventory (Olivier and Berdowski, 2001) and from various regional emissions inventories as described by Nassar et al. (2009). Anthropogenic emissions in Asia are from the Streets 2000 inventory (Streets et al., 2003), with anthropogenic CO emissions in China updated by the Streets 2001 inventory (Streets et al., 2006). These base inventory emissions are scaled to our 2005 simulation year, following van Donkelaar et al. (2008). The 2005 anthropogenic emission for Asia (0–60°N, 65–150°E) is 10.1 Tg N/yr for NO_x and 271.9 Tg/yr for CO. Biofuel emissions follow Yevich and Logan (2003). Biogenic VOCs emissions are based on the Model of Emissions of Gases and Aerosol from Nature (MEGAN) inventory (Guenther et al., 2006). Biomass burning emissions are based on the Global Fire Emissions Database version 2 (GFEDv2), which resolves interannual variability (van der Werf et al., 2006). The monthly GFEDv2 emissions were resampled to an 8-day time step using MODIS fire hot spots (Giglio et al., 2003; Nassar et al., 2009).

Lightning NO emissions in GEOS-Chem are calculated locally in deep convection events with the scheme of Price and Rind (1992) that links flash rates to convective cloud top heights. The spatial distribution of lightning averaged over 11 years in the model is constrained to match the climatological (11-year) satellite observations of lightning flash rates from the Optical Transient Detector / Lightning Imaging Sensor (OTD-LIS) High Resolution Monthly Climatology (HRMC v2.2) product by applying a local monthly rescaling factor (Sauvage et al., 2007; Nassaret al., 2009). The interannual variability of the lightning NO_x emissions as represented by the model is retained, with approximately 6 Tg N yr⁻¹ released globally (Sauvage et al., 2007; Nassar et al., 2009). The vertical distribution of lightning NO_x emissions follows Pickering et al. (1998), with most of NO_x in the upper troposphere (55–75% above 8 km). The model uses the linearized stratospheric O₃ chemistry scheme (Linoz) of McLinden et al. (2000). Liu et al. (2015⁴) previously suggested that Linoz

in a CTM driven with the GEOS-4 and GEOS-5 assimilated meteorological fields yields reasonable impacts of cross-tropopause transport on the lower troposphere.

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Figure Captions

Figure 1. Locations of Beijing (39.8°N, 116.18°E) and three other ozonesonde stations in north China during the second phase of the TAPTO-China campaign in April-May 2005. Surface topography is shown as color image.

Figure 2. Tropospheric O₃ source regions for tagged O₃ simulations.

Figure 3. GEOS-4 (left panels) and GEOS-5 (right panels) horizontal wind vectors near the surface (lower panels) and ~870hPa (upper panels) in East Asia during April-May 2005. Also shown as a color image are the O₃ concentrations (ppbv) simulated by GEOS-Chem driven with GEOS-4 and GEOS-5 meteorological data sets. Values are averages over April-May. White dots denote the locations of four ozonesonde stations (see Figure 1).

Figure 4. a) Mean vertical profiles of O₃ mixing ratios (ppbv) over Beijing averaged over four days (May 1, May 3, May 11, and May 15) when both ozonesonde (black solid line) and MOZAIC aircraft (red dashed line) measurements were conducted. b) Mean vertical profiles of O₃ mixing ratios observed by ozonesonde and MOZAIC aircraft during April-May 2005 (solid black line), in comparison with GEOS-Chem (blue dashed line) and GMI (orange dotted line) simulations driven by the GEOS-4 meteorological fields. Daily model output was sampled at the time and location of ozonesonde and aircraft measurements. c) Same as b), except that GEOS-Chem and GMI were driven by the GEOS-5 and MERRA meteorological fields, respectively.

Figure 5. Time-height cross-sections of lower-tropospheric O₃ mixing ratios (ppbv), as observed by ozonesonde and MOZAIC aircraft in comparison with GEOS-Chem simulations

driven by the GEOS-4 and GEOS-5 meteorological fields, at Beijing during April-May 2005. Model daily outputs are sampled in the gridbox nearest to Beijing for the dates (vertical lines) of ozonesonde soundings.

Figure 6. Major sources contributing to O₃ (0-6km) over Beijing, as simulated by GEOS-Chem driven by the GEOS-4 meteorological fields during April-May, 2005. The plots show time-height cross-sections of concentrations (ppbv, left panels) and percentages (% , right panels) of tagged O₃ produced in Asian, African, European and North American troposphere, as well as O₃ transported from the stratosphere. Model daily outputs are sampled at the location and dates (vertical lines) of ozonesonde soundings and/or aircraft measurements. Note the different color scales on color bars.

Figure 7. Decreases in the lower-tropospheric O₃ concentrations (ppbv), as simulated by GEOS-Chem driven by the GEOS-4 (left panels) and GEOS-5 (right panels) meteorological fields, when Asian, European, North American fossil fuel emissions, biomass burning emissions, or lightning NO_x emissions were suppressed, respectively, relative to their standard simulations for April-May, 2005. Note the different scales on color bars. Colors are saturated when values are out of range.

Figure 8. Timeseries of O₃ concentrations (ppbv, red line) and temperature (black line) at 878 hPa over Beijing in the GEOS-Chem model driven by the GEOS-4 meteorological fields. Also shown in the panel are simulated concentrations (ppbv) of tagged O₃ produced in the Asian (AS, grey line), African (AF, purple line), European (EU, orange line), and North American (NA, blue line) troposphere, and O₃ transported from the stratosphere (Strat, green line). Vertical lines indicate dates with ozonesonde soundings and/or aircraft measurements.

Figure 9. Vertical profiles of O₃ mixing ratios (ppbv) over Beijing from ozonesonde (black solid line) and MOZAIC aircraft (blue solid line) measurements on May 3, 2005, in comparison with those simulated by GEOS-Chem/GEOS-4 (green dashed line) and GMI/GEOS-4 (blue dotted line). Sonde-observed RH (%) and temperature (°C) are shown as red solid line and purple solid line, respectively. Decreases in O₃ concentrations (ppbv) when Asian fossil fuel or lightning NO_x emissions were suppressed, relative to the standard simulation, are shown as orange and purple dot-dashed lines, respectively. O₃ transported down from the stratosphere (ppbv) is shown as pink dot-dashed line.

Figure 10. Five-day back trajectories arriving at the altitudes of 150m (green line), 1400m (blue line), and 2400m (red line), respectively, over Beijing at 1400LT on May 3, 2005. AGL denotes the altitudes above ground level. The ozonesonde station is 34m asl.

Figure 11. Average CO concentrations (ppbv, left panels) and O₃ concentrations (ppbv, right panels) in the lower troposphere (797hPa, top panels; 878hPa, middle panels; 942 bottom panels) over East Asia on May 2, 2005, as simulated by GEOS-Chem driven by the GEOS-4 meteorological fields. Dots denote the locations of four ozonesonde stations shown in Figure 1. Arrows are wind vectors. Note the different arrow scales for wind vectors at different levels.

Figure 12. Same as Figure 11, but for May 3, 2005.

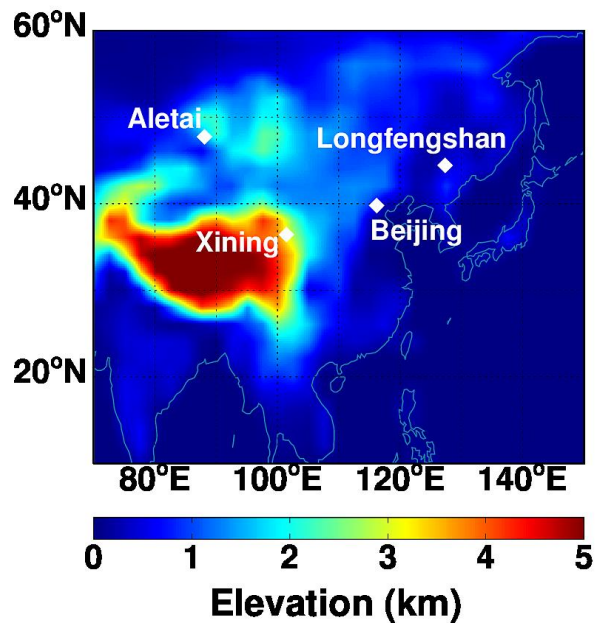


Figure 1. Locations of Beijing (39.8°N , 116.18°E) and three other ozonesonde stations in north China during the second phase of the TAPTO-China campaign in April-May 2005. Surface topography is shown as color image.

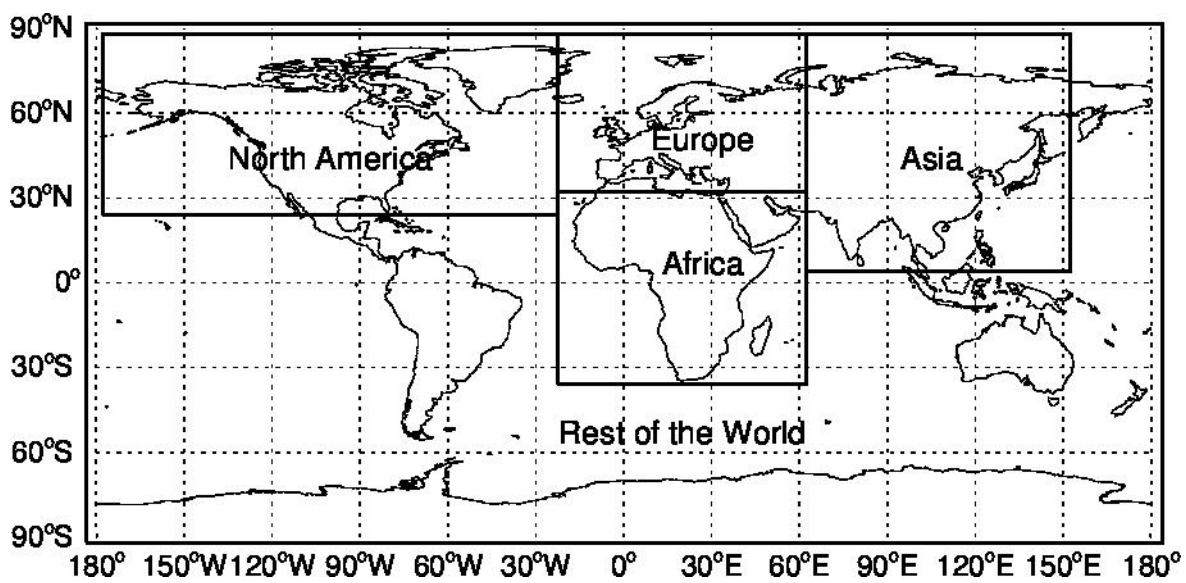


Figure 2. Tropospheric O₃ source regions for tagged O₃ simulations.

GEOS-Chem, April-May

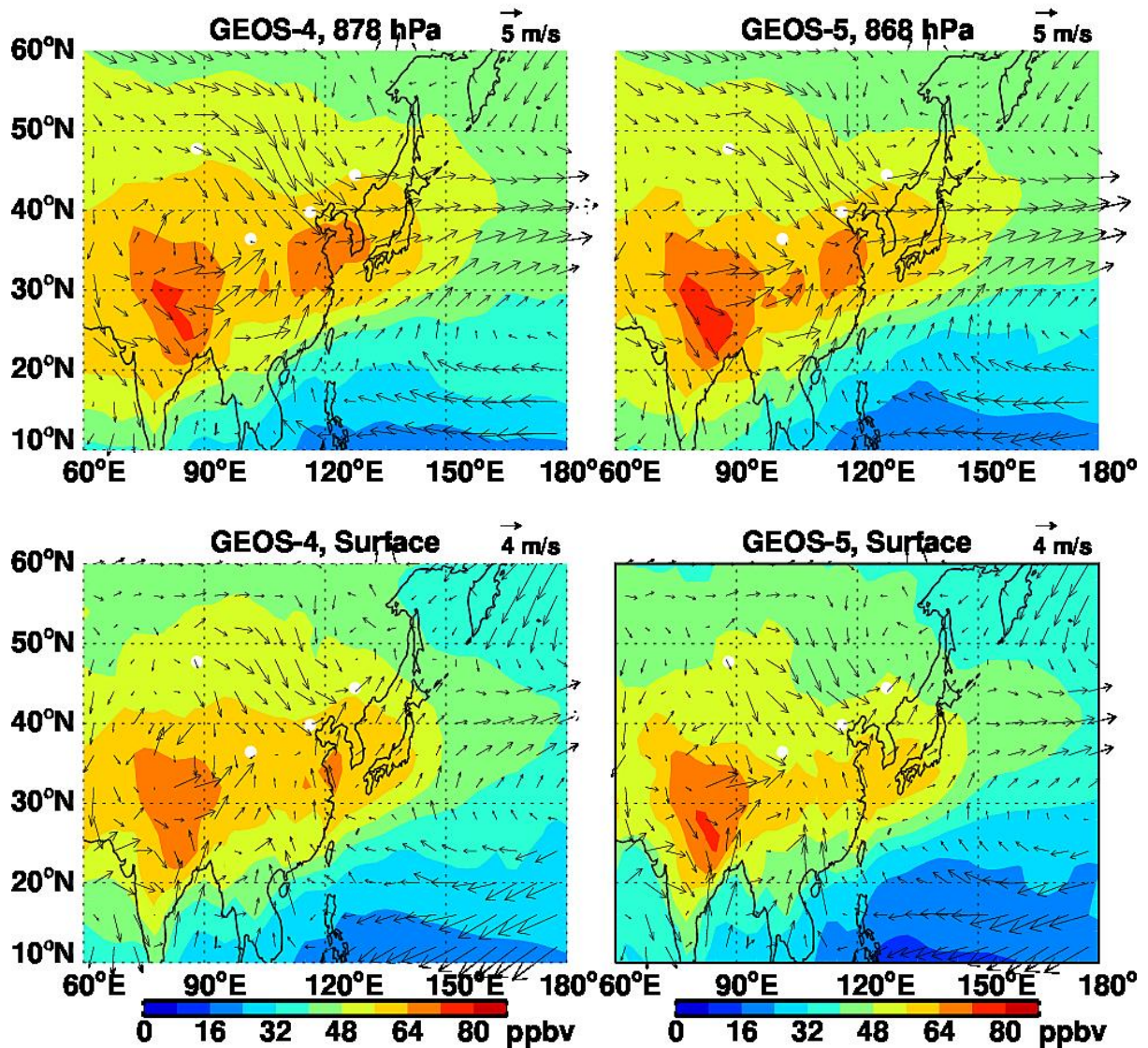


Figure 3. GEOS-4 (left panels) and GEOS-5 (right panels) horizontal wind vectors near the surface (lower panels) and ~870hPa (upper panels) in East Asia during April-May 2005. Also shown as a color image are the O₃ concentrations (ppbv) simulated by GEOS-Chem driven with GEOS-4 and GEOS-5 meteorological data sets. Values are averages over April-May. White dots denote the locations of four ozonesonde stations (see Figure 1).

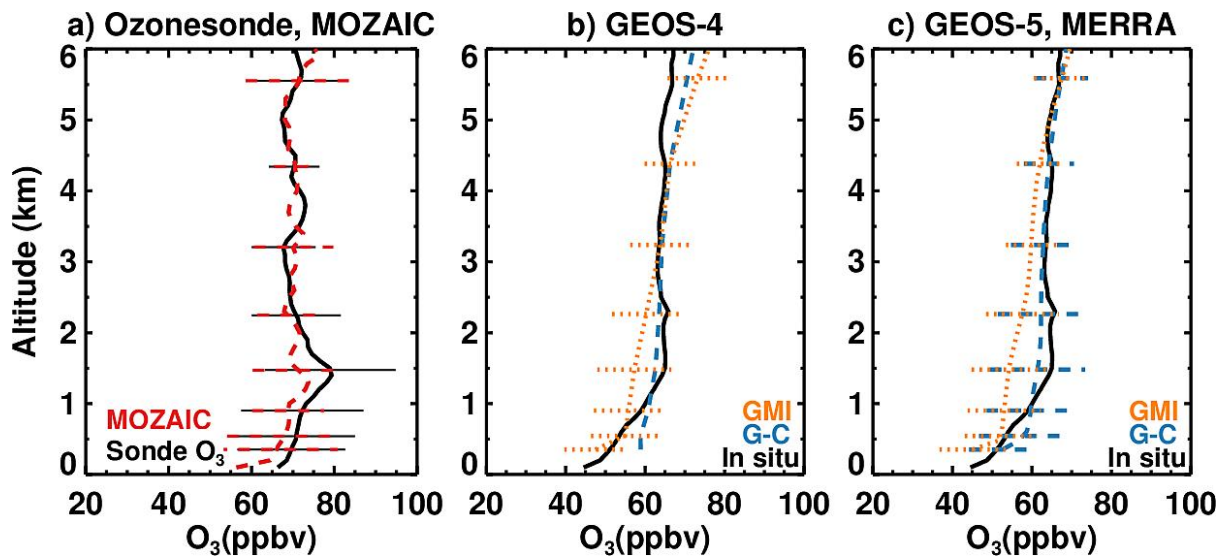


Figure 4. a) Mean vertical profiles of O_3 mixing ratios (ppbv) over Beijing averaged over four days (May 1, May 3, May 11, and May 15) when both ozonesonde (black solid line) and MOZAIC aircraft (red dashed line) measurements were conducted. b) Mean vertical profiles of O_3 mixing ratios observed by ozonesonde and MOZAIC aircraft during April-May 2005 (solid black line), in comparison with GEOS-Chem (blue dashed line) and GMI (orange dotted line) simulations driven by the GEOS-4 meteorological fields. Daily model output was sampled at the time and location of ozonesonde and aircraft measurements. c) Same as b), except that GEOS-Chem and GMI were driven by the GEOS-5 and MERRA meteorological fields, respectively.

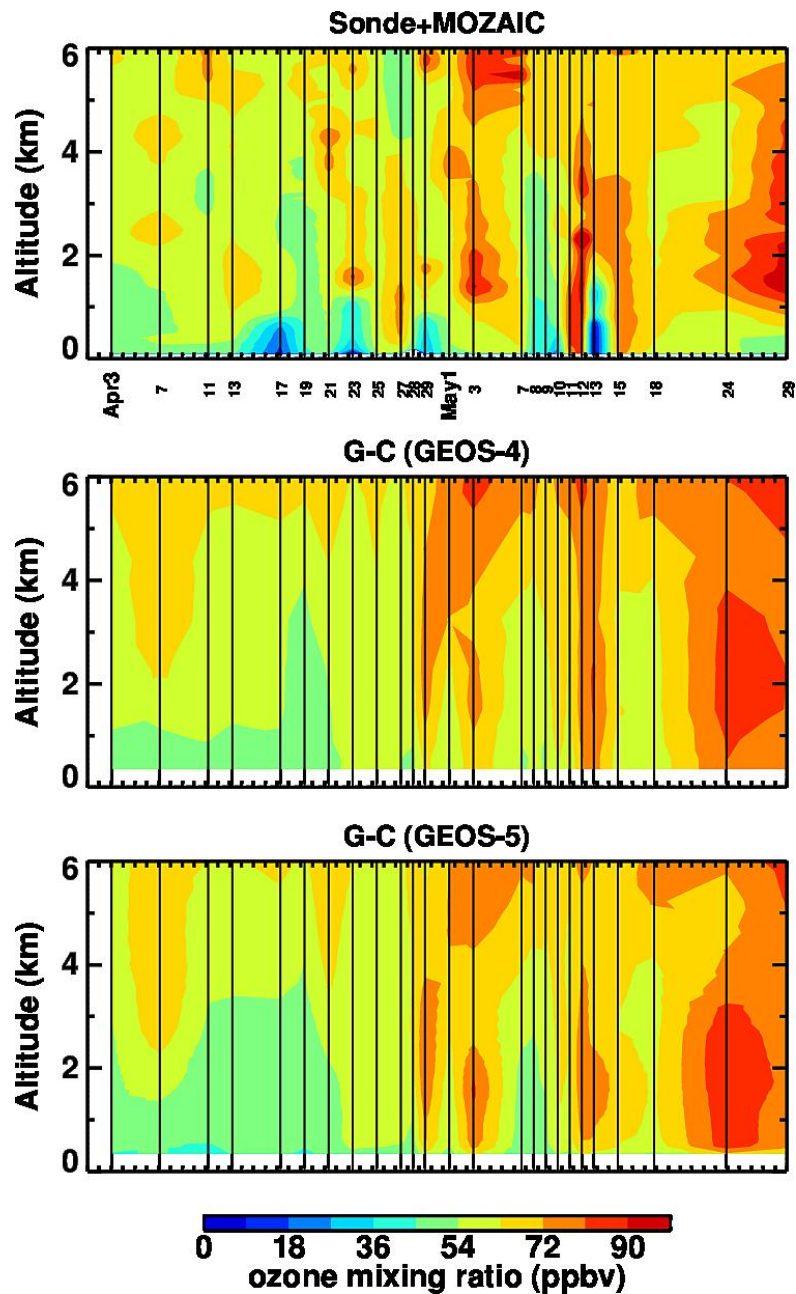


Figure 5. Time-height cross-sections of lower-tropospheric O₃ mixing ratios (ppbv), as observed by ozonesonde and MOZAIC aircraft in comparison with GEOS-Chem simulations driven by the GEOS-4 and GEOS-5 meteorological fields, at Beijing during April-May 2005. Model daily outputs are sampled in the gridbox nearest to Beijing for the dates (vertical lines) of ozonesonde soundings.

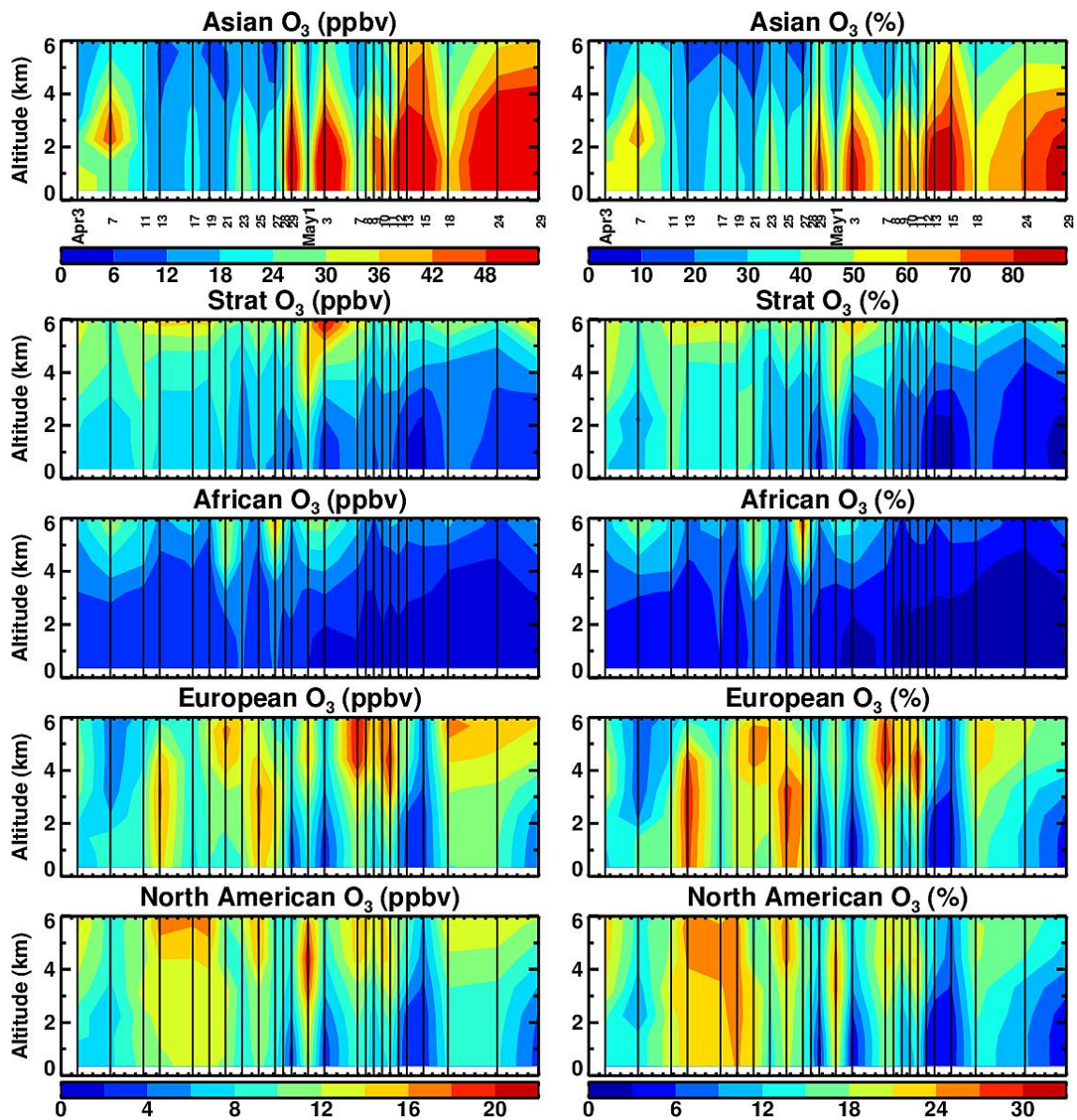


Figure 6. Major sources contributing to O₃ (0-6km) over Beijing, as simulated by GEOS-Chem driven by the GEOS-4 meteorological fields during April-May, 2005. The plots show time-height cross-sections of concentrations (ppbv, left panels) and percentages (% , right panels) of tagged O₃ produced in Asian, African, European and North American troposphere, as well as O₃ transported from the stratosphere. Model daily outputs are sampled at the location and dates (vertical lines) of ozonesonde soundings and/or aircraft measurements. Note the different color scales on color bars.

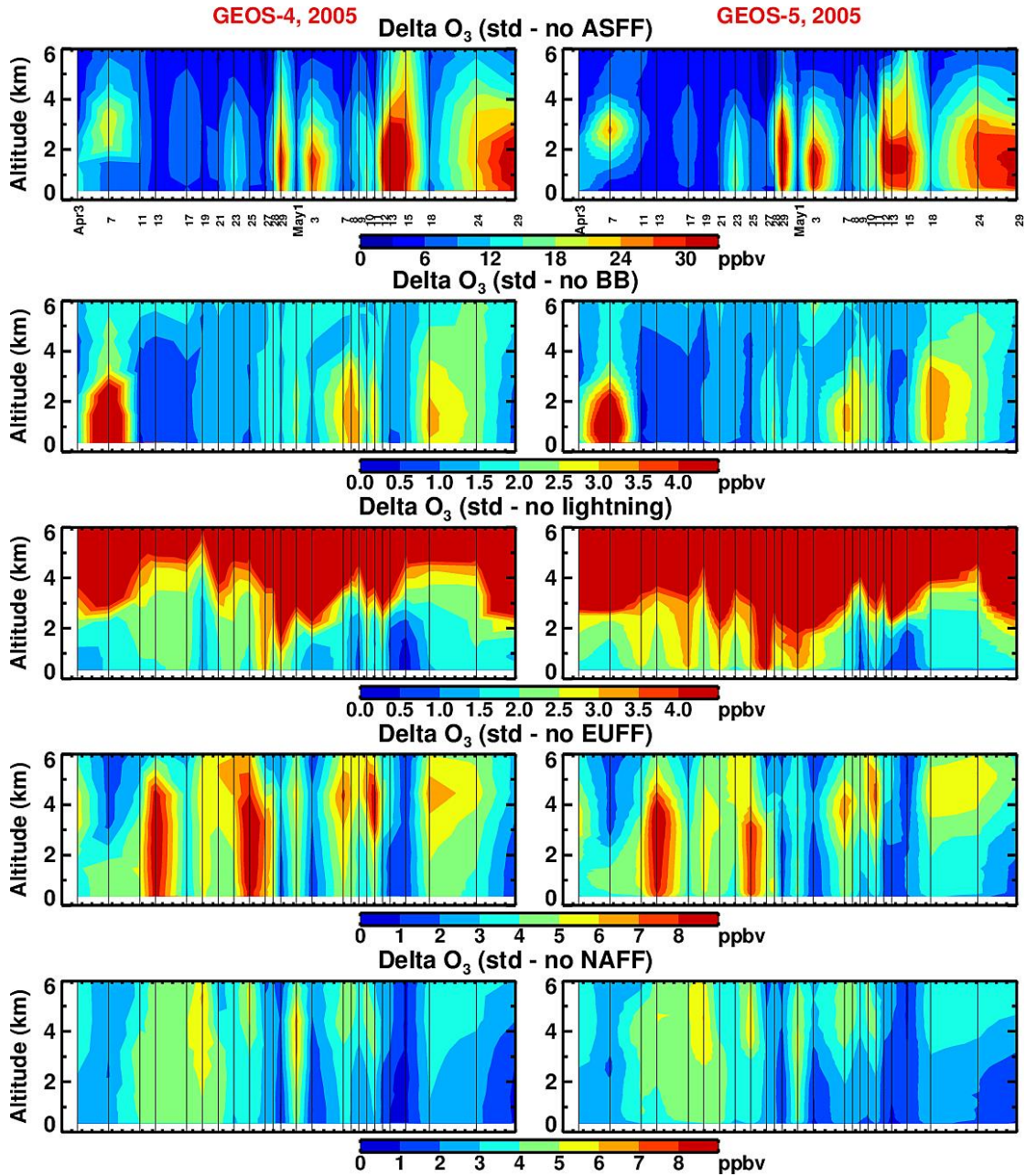


Figure 7. Decreases in the lower-tropospheric O₃ concentrations (ppbv), as simulated by GEOS-Chem driven by the GEOS-4 (left panels) and GEOS-5 (right panels) meteorological fields, when Asian, European, North American fossil fuel emissions, biomass burning emissions, or lightning NO_x emissions were suppressed, respectively, relative to their standard simulations for April-May, 2005. Note the different scales on color bars. Colors are saturated when values are out of range.

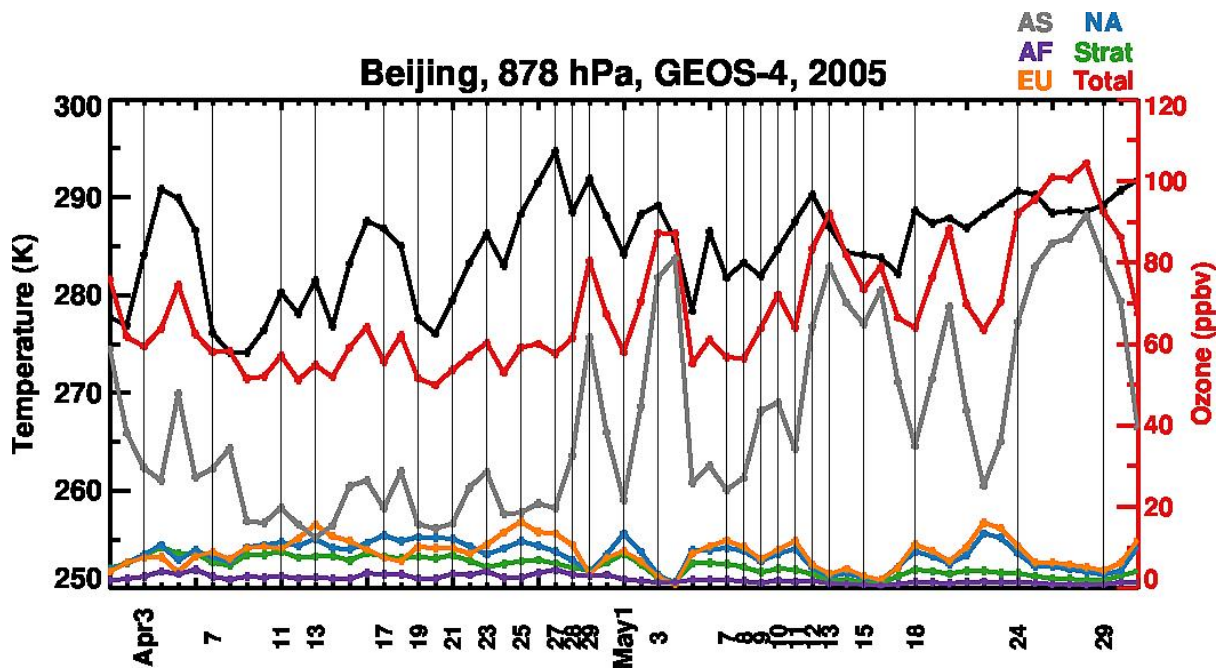


Figure 8. Timeseries of O₃ concentrations (ppbv, red line) and temperature (black line) at 878 hPa over Beijing in the GEOS-Chem model driven by the GEOS-4 meteorological fields. Also shown in the panel are simulated concentrations (ppbv) of tagged O₃ produced in the Asian (AS, grey line), African (AF, purple line), European (EU, orange line), and North American (NA, blue line) troposphere, and O₃ transported from the stratosphere (Strat, green line). Vertical lines indicate dates with ozonesonde soundings and/or aircraft measurements.

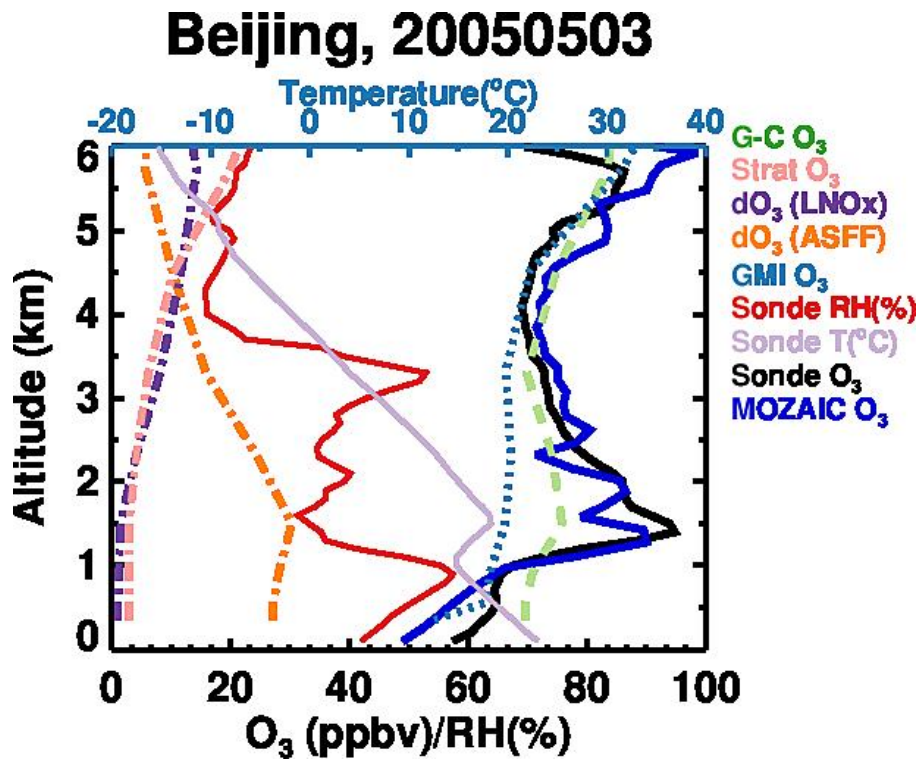


Figure 9. Vertical profiles of O₃ mixing ratios (ppbv) over Beijing from ozonesonde (black solid line) and MOZAIC aircraft (blue solid line) measurements on May 3, 2005, in comparison with those simulated by GEOS-Chem/GEOS-4 (green dashed line) and GMI/GEOS-4 (blue dotted line). Sonde-observed RH (%) and temperature (°C) are shown as red solid line and purple solid line, respectively. Decreases in O₃ concentrations (ppbv) when Asian fossil fuel or lightning NO_x emissions were suppressed, relative to the standard simulation, are shown as orange and purple dot-dashed lines, respectively. O₃ transported down from the stratosphere (ppbv) is shown as pink dot-dashed line.

NOAA HYSPLIT MODEL
 Backward trajectories ending at 0600 UTC 03 May 05
 CDC1 Meteorological Data

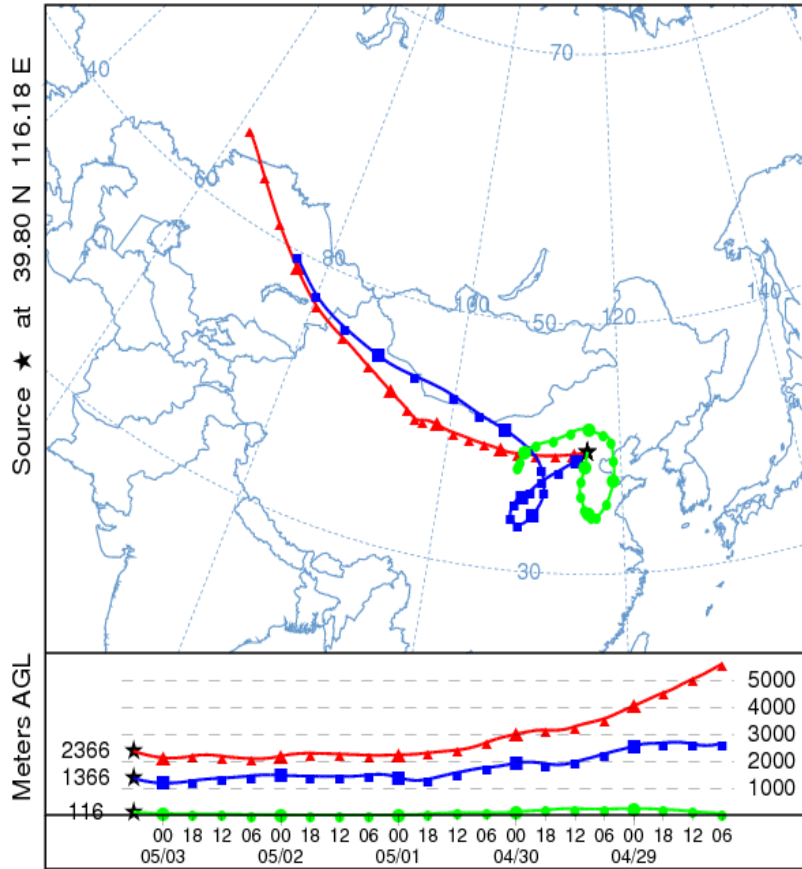


Figure 10. Five-day back trajectories arriving at the altitudes of 150m (green line), 1400m (blue line), and 2400m (red line), respectively, over Beijing at 1400LT on May 3, 2005. AGL denotes the altitudes above ground level. The ozonesonde station is 34m asl.

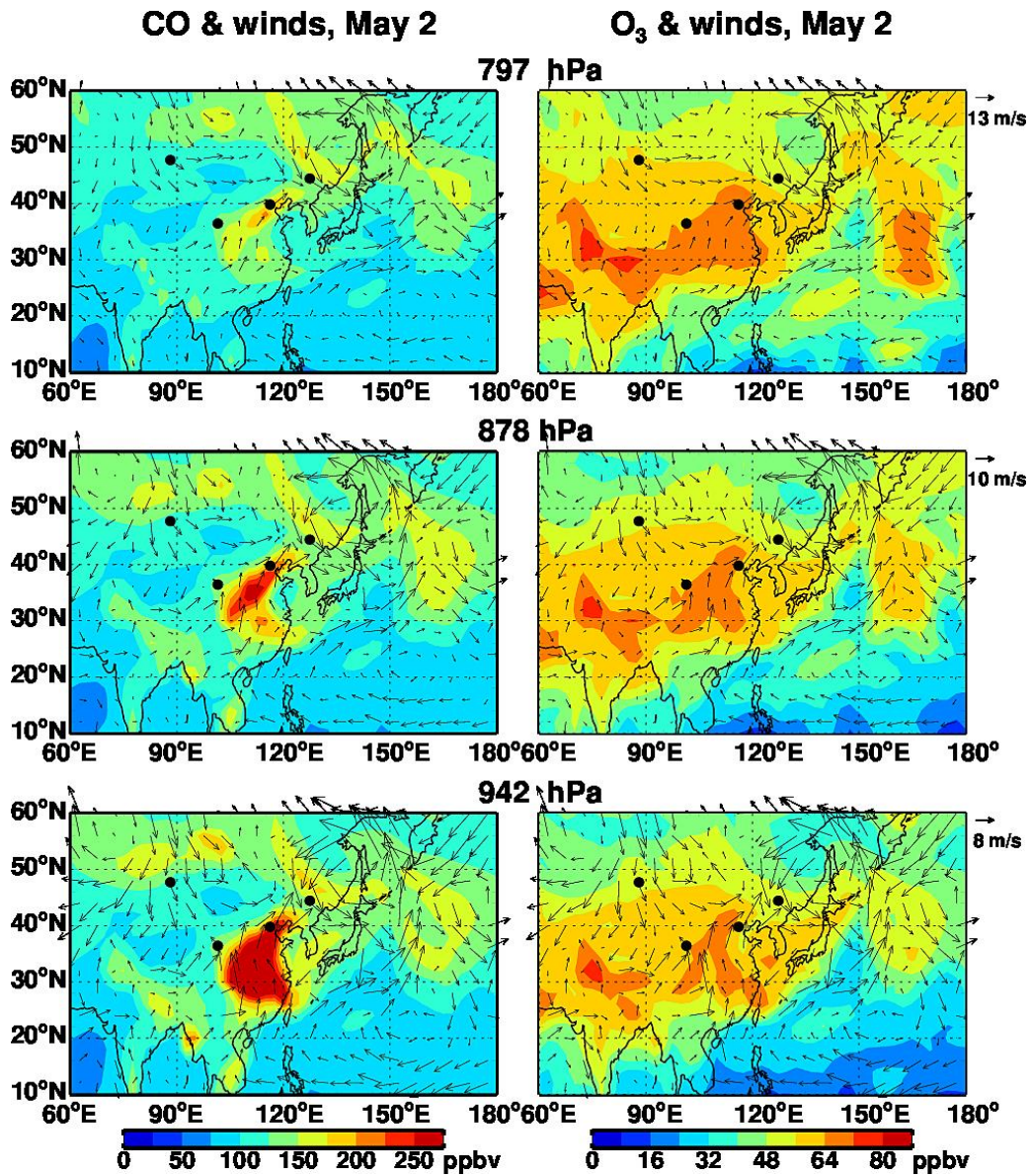


Figure 11. Average CO concentrations (ppbv, left panels) and O₃ concentrations (ppbv, right panels) in the lower troposphere (797hPa, top panels; 878hPa, middle panels; 942 bottom panels) over East Asia on May 2, 2005, as simulated by GEOS-Chem driven by the GEOS-4 meteorological fields. Dots denote the locations of four ozonesonde stations shown in Figure 1. Arrows are wind vectors. Note the different arrow scales for wind vectors at different levels.

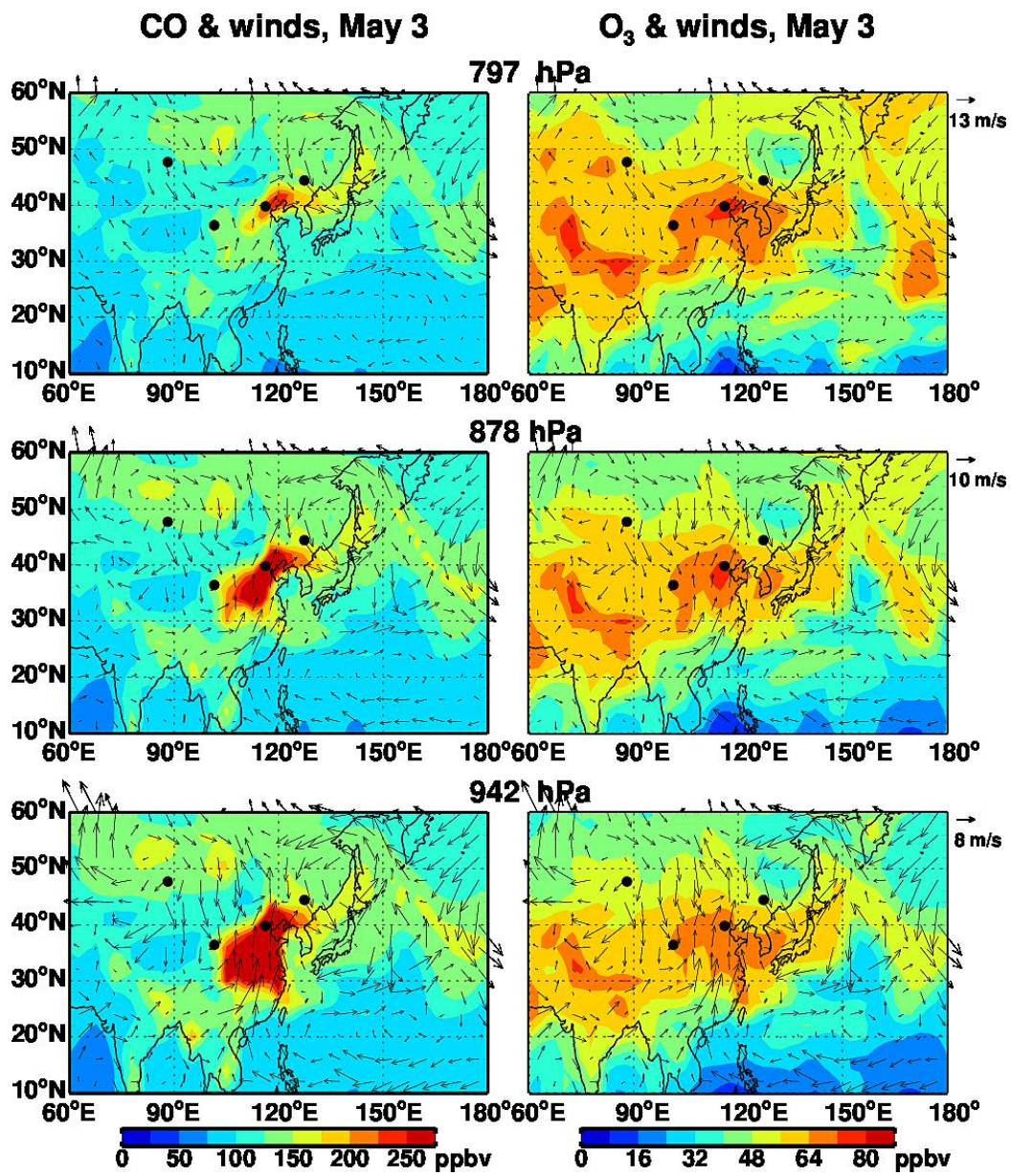


Figure 12. Same as Figure 11, but for May 3, 2005.