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Contributions of regional and intercontinental transport to surface ozone in Tokyo

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Abstract

Japan lies downwind of the Asian continent and for much of the year air quality is directly influenced by emissions of ozone precursors over these heavily-populated and rapidly-industrializing regions. This study examines the extent to which oxidant transport from regional and distant anthropogenic sources influences air quality in Japan in springtime, when these contributions are largest. We find that European and North American contributions to surface ozone over Japan in spring are persistent, averaging 3.5 ± 1.1 ppb and 2.8 ± 0.5 ppb respectively, and are greatest in cold continental outflow conditions following the passage of cold fronts. Contributions from China are larger, 4.0 ± 2.8 ppb, and more variable, as expected for a closer source region, and are generally highest near cold fronts preceding the influence of more distant sources. The stratosphere provides a varying but ever-present background of ozone of about 11.2 ± 2.5 ppb during spring. Local sources over Japan and Korea have a relatively small impact on mean ozone, 2.4 ± 7.6 ppb, but this masks a strong diurnal signal, and local sources clearly dominate during episodes of high daytime ozone. By examining the meteorological mechanisms that favour transport from different source regions, we demonstrate that while maximum foreign influence generally does not occur at the same time as the greatest buildup of oxidants from local sources, it retains a significant influence under these conditions. It is thus clear that while meteorological boundaries provide some protection from foreign influence during oxidant outbreaks in Tokyo, these distant sources still make a substantial contribution to exceedance of the Japanese ozone air quality standard in springtime.

1 Introduction

Air pollution is now recognized as a global issue occurring over hemispheric scales through the intercontinental transport of air pollutants such as tropospheric ozone (O_3), aerosols, mercury, and persistent organic pollutants which have atmospheric lifetimes

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of greater than a week (Akimoto, 2003; Holloway et al., 2003; TF-HTAP, 2007). Rapid industrialization in China, India and other Asian countries has involved increased energy consumption, affecting regional-scale air quality in Asia and beyond now and into the future (Prather et al., 2003; Dentener et al., 2006). Of particular concern is surface ozone, the abundance of which has increased to 40–50 ppb in remote continental areas, a level at which it seriously affects air quality in many parts of the northern hemisphere (Pochanart et al., 2001, 2002; Lin et al., 2001; Fiore et al., 2002). Regional and intercontinental transport affect surface air quality in downwind areas (Jacob et al., 1999; Wilkening et al., 2000; Wild and Akimoto, 2001; Fiore et al., 2002; Jaffe et al., 2003), particularly at mid-latitudes (Berntsen et al., 1999), and also affect the oxidizing capacity of the atmosphere (Thompson, 1992) and regional climate (Berntsen et al., 1996; Mickley et al., 1999). It is now realized that intercontinental transport may affect local O_3 air quality, adding to the effects of regional transboundary transport and local photochemical formation (TF-HTAP, 2007; Fiore et al., 2009). Previous atmospheric chemistry modeling studies have shown that increased precursor emissions in Asia may be sufficient to offset reductions in American emissions and may thereby counteract the beneficial effects of air quality controls in the US (Jacob et al., 1999; Yienger et al., 2000; Heald et al., 2003). In addition, regional increases in surface background O_3 and episodic exceedances of O_3 air quality standards are likely to damage agriculture and human health in China and East Asia, leading to economic damage (e.g., Mauzerall and Wang, 2001; Wang and Mauzerall, 2004) as many developed countries have already experienced (Lippmann , 1991). Thus, a quantitative understanding of the contributions of inter- and intra-continental transport may be as important for future air quality policy-making as an understanding of the effects of local emission controls (Wilkening, 2001; Fiore et al., 2003; Holloway et al., 2003).

Japan lies directly downwind of the Asian continent and is particularly susceptible to emissions from China and other Asian countries. Increases in surface O_3 have been reported over Japan in the past decade (Lee et al., 1998; Ohara and Sakata, 2003; Naja and Akimoto, 2004). This is of particular concern in the metropolitan area of Tokyo,

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where measurements of total photochemical oxidants (O_x, predominantly O_3) have increased since the 1980s despite strong decreases in the main precursors, nitrogen oxides (NO_x=NO + NO_2) and non-methane hydrocarbons (NMHCs) (Tokyo Metropolitan Government, 2005). It is not clear to what extent increased emissions of O_3 precursors over China (Irie et al., 2005; Ohara et al., 2007) have influenced this trend, but it is likely that long-range transport of O_3 from the Asian continent has played a role. However, a more complete attribution of these observed changes depends on a good understanding of the factors affecting O_3 over the region, and on a reliable quantification of the roles of inter- and intra-continental transport, stratospheric influence, and in situ formation in controlling O_3, and an understanding of how these factors are controlled by meteorological processes.

The composition of air arriving over Japan is heavily influenced by regional meteorology. Direct transport of polluted air from the Asian mainland is strongest in springtime and is dominated by the passage of mid-latitude cyclones (Kaneyasu et al., 2000; Bey et al., 2001; Wild et al., 2003; Tanimoto et al., 2005). High levels of pollutants are found in the boundary layer following these frontal systems (Carmichael et al., 1998; Liu et al., 2003; Liang et al., 2004; Sawa et al., 2007), and pollutants lifted by convection and warm conveyor belts in these systems may be rapidly transported across the Pacific (Bey et al., 2001; Stohl, 2001; Heald et al., 2003; Miyazaki et al., 2003) and subsequently detected over North America (Jaffe et al., 2003; Jaeglé et al., 2003). Influence from more distant sources such as Europe may affect Japan during periods of post-frontal outflow from Asia (e.g., Liu et al., 2003; Lin et al., 2010), but the magnitude and timing of these events remains uncertain. Photochemical production of O_3 is greatest in summertime, but over Japan exceedance of urban air quality standards is more common in spring, as the prevailing westerly flow brings continental air with much higher levels of O_3 than is present in the tropical marine air from the Western Pacific that dominates in summer (e.g., Tanimoto et al., 2005; Yamaji et al., 2006). Continental sources may thus affect air quality attainment in Japanese cities, but the extent of their influence is not known.

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This study aims to determine how local, regional and distant sources contribute to surface O_3 over Japan in springtime and the extent to which they influence episodes of high O_3 over the Tokyo metropolitan area. We use observations of O_3 and CO from surface measurement sites in Japan and over the Western Pacific in spring 2001 to evaluate a global chemistry-transport model in Sect. 2, and then use the model to determine the contribution of selected precursor emission sources to O_3 over Japan during this period. In Sect. 3 we focus on the mean contribution from these sources and on their influence during key periods when O_3 air quality standards are exceeded. We then examine the meteorological mechanisms that bring this air to Japan in Sect. 4, focusing on the pathways and timing of transport from the different source regions from a climatological perspective and during transport episodes of particular interest. We conclude by summarizing the influence that regional and intercontinental transport has on springtime air quality in Japan.

2 Model and evaluation against observations

This study uses the Frontier Research System for Global Change (FRSGC) version of the University of California, Irvine (UCI) global chemistry-transport model (CTM) (Wild and Prather, 2000) with the configuration described in Wild et al. (2003). The CTM is driven with pieced-forecast meteorological fields generated with the European Centre for Medium-Range Weather Forecasts Integrated Forecast System (ECMWF-IFS) at a spectral resolution of T159L40 at 3-h intervals, and these data are used here at T63 ($1.9^\circ \times 1.9^\circ$) and T21 ($5.6^\circ \times 5.6^\circ$) resolution. The model was initialized in January 2000 and run for 16 months at T21 resolution, allowing a 1-yr spin-up and then covering winter and spring 2001. The period from January to April 2001 was repeated at the higher T63 resolution. The spring 2001 period was chosen to coincide with the NASA Transport and Chemical Evolution over the Pacific (TRACE-P) measurement campaign over the Western Pacific (Jacob et al., 2003), and previous studies using the model have been evaluated against observational data from this campaign (Wild et al., 2003).

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Global industrial and fossil fuel emissions of O_3 precursors used in this study are taken from version 2 of the EDGAR database (Olivier et al., 1996), and emissions for East Asia are updated from Streets et al. (2003); these and other emissions are described in more detail in Wild et al. (2003).

The performance of the model during spring 2001 is evaluated with hourly observational data for surface O_3 from 10 measurement sites in the Acid Deposition Monitoring Network in East Asia (EANET) (Network Center for EANET, 2005) and with O_3 and CO from four World Meteorological Organization/Global Atmosphere Watch (WMO/GAW) stations (Tsutsumi et al., 2006). The location of these measurement sites is shown in Fig. 1 and detailed in Table 1. The sites are loosely classified here as remote, rural or urban based on their chemical environment. Eight clean coastal or mountain sites are categorized as remote, three sites showing strong pollution signatures are designated as urban, and the remaining three sites showing some influence from local sources are denoted as rural.

We first examine the model performance at remote sites which provide an effective test of the ECMWF-IFS meteorology and the ability of the CTM to correctly represent the chemical environment in clean, background conditions. Figure 2 shows the hourly timeseries of O_3 and CO at Minamitorishima, a remote island lying almost 2000 km southeast of Tokyo. The prevailing influence in early spring is from continental air masses that have crossed Asia, but this flow is increasingly punctuated by the arrival of clean, marine air masses from the Central Pacific. The variability in O_3 and CO in springtime is largely driven by the interplay between these air masses, and the location and timing of the meteorological features controlling this variability are captured very well by the ECMWF-IFS model. Surface O_3 in continental air averages about 50 ppb, and in marine air averages about 15 ppb, and these abundances are reproduced well by the CTM, suggesting that the background chemical environments over the Western Pacific in spring can be represented well. The variability in CO is smaller, but frequent episodes of high CO reveal the passage of cold frontal systems which sweep away the clean, marine air and bring CO directly from the Asian continent, as previous studies

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have shown (Liu et al., 2003; Sawa et al., 2007). The abundance of CO in these environments is captured well by the CTM. The results are not very sensitive to model resolution at this location, although the position of meteorological features is better resolved at T63 where there is less spatial averaging of the input meteorological data, and the model bias is consequently reduced.

Polluted, continental conditions are more challenging to model but provide a more critical test of emissions, chemistry and deposition processes in the CTM. Figure 3 shows the time series of afternoon-mean surface O_3 at Tsukuba, a suburban site about 50 km northeast of Tokyo. There is a strong diurnal variation in the observations, with very low nighttime O_3 driven by both surface deposition and direct chemical removal by freshly-emitted NO. Fast chemical removal in the stable nocturnal boundary layer is not reproduced well at the coarse model resolutions used here, but is notably better at T63 resolution (180 km scale) than at T21 (550 km scale) as the artificial mixing of fresh pollutants to the model grid scale is less extensive. The Tsukuba site lies in the same model grid square as Tokyo at both resolutions, but cannot be considered representative of the whole urban area. To minimize the influence of nighttime removal we compare afternoon concentrations (12:00–16:00 h local time) which are dominated by photochemical production of O_3. These values are matched more closely by the CTM, and while there is still overestimation of all but the highest daytime levels in February and March, the main features and magnitude of urban O_3 are reproduced well in April.

The performance of the model at other locations is shown in Fig. 4, and a statistical summary of the comparisons for the whole February–April 2001 period is given in Table 2. Where observational data are missing due to instrument failure or calibration errors, model data are also removed so that the sampling is identical, and the number of observations used is given in Table 2. Mean values, mean bias and root-mean-square error (RMSE) are presented for both T21 and T63 simulations. The mean bias and RMSE are generally less than 10 ppb for the more remote measurement sites, but are substantially greater at rural and urban sites where the efficiency of nighttime

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removal is not captured. The remote site at Yonagunijima lies close to Taiwan and is strongly influenced by urban emissions from Taipei at the coarse model resolutions used here; similarly Oki and the coastal sites of Yusuhara, Sadoseki and Ryori are dominated by marine air flows but are artificially influenced by Japanese industrial regions in the model. Tappi shows little variability in O_3 in air from the continent in February, but experiences high levels of O_3 from Japanese sources in April as the prevailing flow changes. Island sites south of Japan such as Ogasawara, Hedo and Yonagunijima show the higher variability in O_3 characteristic of frontal influence, as seen at Minamitorishima, although there is a tendency to overestimate O_3 in continental air, particularly at T21 resolution. Comparison with CO at the WMO/GAW stations, summarized in Table 3, presents a similar picture, with synoptic features captured well, but there is a small negative mean bias, and CO is underestimated during pollution episodes, contributing to the higher RMS error.

The model run at T63 (1.9°) resolution generally shows smaller mean biases and RMS errors than the T21 (5.6°) run as meteorological features are better resolved, see Table 2. Mean O_3 levels are lower at T63 reflecting an overestimation of regional O_3 production and background continental O_3 inherent at the coarser T21 model resolution (e.g., Wild and Prather, 2006), and the variability is captured better. The performance is poorer at only a few locations, notably at Tappi where the site is less representative of the model grid box at T63 than at T21, and at Ogasawara, where overestimated O_3 in continental air is compensated at T21 by too great a marine influence in April. Note that at T21 resolution some of the sites lie in the same model grid box (e.g., Ijira, Oki and Yusuhara), and therefore show the same variation; these locations lie in different grid boxes at T63. The following sections of this study focus on model simulations at T63 resolution only.

Two of the sites considered here, Oki and Happo, lie upwind of the major Japanese source regions, and may intercept outflow from the Asian mainland before it arrives over Tokyo. Both sites are strongly influenced by pollution from China, and O_3 from this source is transported efficiently in spring (Berntsen et al., 1996; Wild and Akimoto, 2001).

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The sampling environment at these locations is very different, and O_3 is overestimated in the marine boundary layer sampled at Oki but underestimated at the mountain site at Happo, see Table 2. However, the variations in O_3 at these sites and at Tsukuba are well matched (the correlation between O_3 data at Oki and Tsukuba is $r^2 = 0.46$ in observations and 0.45 in the model; for Happo and Tsukuba, $r^2 = 0.53$ and 0.52, respectively), demonstrating that background O_3 makes a substantial contribution to the variation in O_3 over Tokyo. The seasonal trend at Oki is also well matched, with an increase in monthly mean O_3 of 15.3 ppb between February and April (14.6 ppb in model), but is overestimated at the higher elevation of Happo (14.3 ppb in observations, 22.3 ppb in model) and underestimated in the more polluted conditions at Tsukuba (25.4 ppb in observations, 15.4 ppb in model). During conditions of westerly flow there is a time lag in major features in CO of about 12 h between Oki and Tsukuba, reflecting the timescales for transport of polluted continental air between the sites.

3 Contributions to surface O_3 and CO over Tokyo

We evaluate the contribution of industrial and fossil fuel sources of O_3 precursors to O_3 over Japan from five independent source regions: local emissions from Japan and Korea (JP), regional emissions from China (CH) and southern Asia (SA), and remote emissions from the major source regions of Europe (EU) and North America (NA), see Fig. 5. In addition, biomass burning sources (BB) are considered for southern Asia where peak emissions occur in the February–April period examined here (Duncan et al., 2003; Streets et al., 2003). These originate from the same region as SA emissions, but the distribution of sources is different, as industrial emissions dominate over India while biomass burning sources are greatest over Thailand and Southeast Asia. Source contributions are evaluated by removing emissions of NO_x, CO and VOC from each region in turn and comparing the resulting simulation with the control run. These perturbation scenarios were initialized in January 2000 and were run until the end of April 2001 at low resolution (T21), as for the control run; the January–April 2001

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period was then repeated at higher resolution (T63), and the results from these runs are described here. This method provides a reasonable estimate of source contributions, although non-linearities in O_3 chemistry prevent a full and accurate attribution of all sources using this method (see, e.g., Wu et al., 2009). Stratosphere-troposphere exchange (STE) is also an important source of O_3 in the northern hemisphere in springtime, and O_3 from this source is diagnosed by accounting for the chemical destruction of O_3 transported from the stratosphere following the method of Roelofs and Lelieveld (1997).

The modeled contribution of different source regions to surface O_3 and CO over the Tokyo region is shown in Figs. 6 and 7. These results are representative of a 180 km grid box covering a much wider region than the Tokyo metropolitan area, and while they do not capture the heterogeneity of chemical environments close to such a major source region, or nighttime removal, the mean photochemical environment is captured sufficiently well to reproduce the day-to-day variability in O_3 shown in Fig. 3. Ozone shows an upward trend over the spring period as photochemistry becomes more active, while CO is at its annual maximum and shows no trend over the period; however these trends are masked by large variability from both local and regional sources. This variability is driven largely by local emissions; for O_3 it is dominated by the photochemical impacts of local NO emissions over Japan, which lead to production during the daytime and destruction at night, while for CO it is driven by local meteorology and transport processes from sources over Japan. The contribution of Japanese sources to O_3 increases from February to April as photochemical activity increases, and there is also an increase for CO, as weakening of the northwesterly winter monsoon flow allows increased influence from local sources. Chinese contributions to O_3 and CO (averaging 4.0 ± 2.8 and 23 ± 13 ppb respectively) are strongly episodic in nature, reflecting regional meteorological patterns, and episodes occur about twice a week on average. The highest impacts reach 13–15 ppb for O_3 and 60–80 ppb for CO, and CO from Chinese sources generally exceeds that from Japanese sources during these episodes. At other times the contribution of Chinese sources is smaller than that from more distant

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sources like Europe and North America. Chinese contributions to O_3 and CO are well correlated ($r^2 = 0.82$), as shown in Fig. 8, highlighting the good relationship between emissions of O_3 precursors and CO and the relatively short timescales for transport to Japan. The correlation is much weaker for other source regions where O_3 destruction and differing transport pathways mask the relationship. The O_3/CO ratio from Chinese sources increases from 0.17 to 0.29 between February and April as photochemical activity increases, similar to that observed over the North Atlantic in outflow from North America (Parrish et al., 1993).

European and North American contributions are less variable, reflecting the greater transport distances from the source regions. European O_3 contributions average 3.5 ± 1.1 ppb and rise from 2.5 to 4.1 ppb between February and April, but the contribution from North America is relatively constant at 2.8 ± 0.5 ppb. Contributions of European and North American sources to CO average 30 and 20 ppb respectively, and the variability is again less for North American sources. The magnitude and trends of these O_3 and CO contributions agree well with a previous study using different emission data (Wild et al., 2004) and North American CO is similar to but slightly larger than that reported by Liang et al. (2004) who found contributions of up to 15 ppb at Ryori, Yonagunijima and Minamitorishima in spring. The contribution of industrial and biomass burning emissions from South and Southeast Asia is small at this latitude, averaging 0.5 and 0.3 ppb for O_3, respectively, and about 4 ppb each for CO. However, there is one episode bringing 15 ppb of CO from biomass burning sources on 1 February. In contrast to these surface sources, the contribution from stratospheric O_3 is large, 11.2 ± 2.5 ppb, and is highly variable from day to day, exceeding 20 ppb on two occasions. The stratospheric contribution is larger in March than in April, indicating that the positive trend in surface O_3 seen over Japan in spring is driven largely by photochemistry, as previous observational studies have suggested (Kajii et al., 1998; Pochanart et al., 1999).

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The monthly contributions of the selected source regions to CO and O_3 over Tokyo in spring 2001 are summarized in Tables 4 and 5. Local sources of CO and O_3 over Japan play an increasingly important role during the spring, as photochemistry picks up and long-range transport from distant source regions weakens. For O_3 we consider only the afternoon hours (12:00–16:00 JST) when photochemistry is most active and the boundary layer is relatively deep to avoid the influence of nighttime removal. Local sources dominate, exceeding 20% in April, but this is only marginally larger than the influence of the stratosphere (18%) and the combined impacts of fossil fuel sources over China, Europe and North America (18%). These contributions are similar to those derived for March to May for the whole of Japan by Nagashima et al. (2010), who found contributions from the stratosphere of 21%, from Japanese sources 22%, and from Chinese sources 12%, slightly greater than found here. It is thus clear that long-range transport makes a large contribution to daytime O_3 over Tokyo on a mean basis throughout the spring.

The source contributions are shown in Fig. 9 as a function of the total surface O_3 to demonstrate their influence during episodes of high O_3 over Tokyo. There is a very high correlation between the Japanese contribution and total surface O_3 ($r^2 = 0.83$) indicating that local sources account for most of the variability in O_3 through their control over the diurnal cycle, and they also account for much of the day-to-day variability ($r^2 = 0.42$) reflecting their control over local photochemistry. Chinese sources show a weaker relationship ($r^2 = 0.15$) but still contribute an average of 5.7 ppb during periods when O_3 exceeds the Japanese 1-hour air quality standard of 60 ppb. European and North American sources both show a weak correlation, and contribute an average of 4.5 ppb and 3.1 ppb to episodes with O_3 exceeding 60 ppb, respectively. These contributions from distant sources are similar to those seen in the summertime boundary layer over the US (Fiore et al., 2002). The stratospheric contribution to surface O_3 lies tightly between 15 and 33% over the spring period, and although there is a notable decline when surface O_3 exceeds 50 ppb, the stratosphere still contributes an average of 12.2 ppb during O_3 episodes.

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During the highest episodes O_3 was observed to exceed 80 ppb at Tsukuba for a total of 48 h over the spring period, compared with 25 h in the model. The contribution of each source region during the modeled exceedance periods is shown in Table 6. The six episodes shown differ substantially in nature. On 12 April (day 101), Japanese sources contributed more than 50% of the O_3 peak, and industrial emissions from Chinese, European and North American sources together accounted for less than 10%. However, during the 20 March (day 79) and 20 April (day 109) episodes Japanese sources account for only about 25% of the O_3, and Chinese sources account for more than 18%. European and North American contributions remain significant during these periods, accounting for almost 10%. Regional trans-boundary transport contributed significantly to the exceedance of air quality standards during these episodes. The remaining three episodes lie between these extremes, with Japanese sources accounting for about one third of O_3 and other industrial sources for 15–20%. The stratospheric source remains a consistent contributor during these episodes, accounting for 8–12 ppb (10–15%). Comparing contributions during episodes with mean afternoon contributions for April, it is clear that while local Japanese sources play a much bigger role than normal during episodes, contributions from distant sources are only marginally smaller than average. This suggests that trans-boundary and intercontinental transport may contribute significantly to O_3 levels over Japan during conditions that favour O_3 build-up, and may thus impact attainment of air quality standards in Japan.

4 Transport pathways and meteorology

The large differences seen between high O_3 episodes suggest that the source contributions are very sensitive to meteorological conditions. We therefore examine how regional meteorology controls the transport of oxidants from different source regions to characterize better the composition of regional "background" air on which Japanese sources build. Meteorological conditions over East Asia in springtime are characterized by a weakening of the Siberian High that drives the northwesterly East Asian

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winter monsoon flow. This cold, dry continental air from over Eurasia meets warmer, more humid air over the Western Pacific, and leads to frequent and active cyclogenesis over the region. The wave cyclones that form travel northeastwards over Japan and typically increase in strength over the early spring period considered here. Continental pollutants may be rapidly swept out over the Western Pacific in the boundary layer following the associated cold fronts (e.g., Carmichael et al., 1998; Liu et al., 2003) or may be lifted into the free troposphere over the Western Pacific in the warm conveyor belt flow ahead of the front (e.g., Kaneyasu et al., 2000; Yienger et al., 2000; Miyazaki et al., 2003). Associated with these systems is a strong jet stream, and significant influx of stratospheric O_3 may occur in dry air descending behind the cold front (e.g., Cooper et al., 2002). The passage of these systems leads to frequent changes in air mass origin over much of the Western Pacific, and there is a gradual transition in this variability as the marine flow strengthens. The meteorology of the region in 2001 is reasonably typical, influenced by a weak La Niña that decayed to neutral conditions in early spring (Fuelberg et al., 2003).

4.1 Meteorological controls on source region contributions

The juxtaposition or coalescence of air masses from different origins can be explored by examining the relationships between the contributions of different sources with time. Figure 10 shows these relationships for CO and O_3 over Japan in February and April 2001. European and North American contributions to CO are well correlated, indicating that they arrive together in relatively well-mixed air masses. This signal is much stronger in March and April than in February, highlighting differences in synoptic conditions. The correlation for O_3 differs somewhat from that of CO, as O_3 production is strongly influenced by meteorological conditions over the source region and by the different timescales for transport. North American O_3 correlates relatively well with stratospheric O_3 suggesting that they arrive by similar pathways following transport and subsidence from the middle and upper troposphere. European O_3 correlates with

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stratospheric O_3 marginally less well, consistent with transport at lower altitudes than North American air masses (Wild et al., 2004). Previous studies have suggested that European CO is mixed with CO from mainland Asian sources in post-frontal boundary layer outflow from China (Liu et al., 2003), but we find European and Chinese contributions are quite strongly anticorrelated in February, and only weakly correlated in April. This suggests that over Japan the timing of major episodes differs substantially; in February the relationship is strongest when European influence lags Chinese influence by about 48–60 h, reflecting changes in air mass origin driven by changing synoptic systems, while in April the relationship is strongest with a lag of about 6 h, likely reflecting Chinese influence in bands immediately behind frontal systems giving way to European influence in the following cold continental outflow.

To determine the governing meteorological mechanisms and the dominant transport patterns that bring air from different source regions to Japan in springtime we derive correlation maps of the hourly time series at Tokyo to mean sea-level pressure over the region and relate these to the prevailing meteorological conditions. Figure 11 shows this correlation map for the seasonal time series of Chinese, European and Japanese influences on Tokyo. The correlation patterns for O_3 and CO are similar for Chinese sources, but the strong diurnal signal in O_3 from Japanese sources and the lower variability from European sources give a weaker response, and we therefore show correlations for CO which highlight the meteorological patterns more clearly. For Chinese sources, the region of anticorrelation north of Japan indicates the climatological location of low pressure systems and associated cold fronts which bring higher influence from the continent to Japan, and the classic cyclonic "comma" shape is clearly visible. Applying a 24-h lag to the time series reveals the climatological position of the low pressure one day earlier, over Korea, and demonstrates that high levels of O_3 from China are associated with the passage of low pressure systems as their attendant frontal systems advance towards Japan. These patterns highlight the start of the North Pacific storm track, and show that transport of Chinese pollutants to Japan is favoured by the movement of low pressure systems northeastwards from the East China Sea.

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The characteristic pattern produced by this analysis is similar to that shown by Liang et al. (2005).

The correlation map for European influence is notably different and indicates that import of European air is favoured when there is higher sea-level pressure over the East China Sea along with low pressure systems northeast of Japan. This pattern is associated with northwesterly flow from Northern Eurasia to Japan, and follows on from northerly flow one day earlier. The climatological pattern represents cold outflow around the eastern side of the high pressure systems that follow the passage of wave cyclones. This is supported by an anticorrelation with humidity ($r^2 = 0.5$, not shown) which indicates that European influence is greatest in dry continental air masses.

The correlation maps for Japanese influence are very different, and the greatest impacts on CO and O_3 are associated with a high pressure anomaly over the northern Pacific and a low over Eastern China. This pattern reflects anticyclonic conditions over Japan governed by slow marine southwesterly flow, and these fair-weather conditions allow substantial build-up of local pollutants in the boundary layer. These conditions are relatively short-lived in spring and are typically swept away by the frontal systems associated with the next cyclone building over Eastern China.

4.2 Case study of inflow to Japan

While climatological patterns provide good evidence for the role of particular meteorological processes in governing transport on a seasonal basis, there is substantial variability between one synoptic system and the next. To demonstrate the key pathways for regional pollutant transport more clearly, we present a case study of the transport of continental Asian pollutants to Japan during 23–26 February 2001. Of the transport episodes we have examined, this case is reasonably typical of transport from China associated with frontal passage. Polluted air from China is transported eastwards over the Pacific, bringing high levels of O_3 (12–14 ppb) to Japan, but is subsequently caught up in a region of cyclogenesis over the East China Sea, see Fig. 12. The passage of this cyclone along the North Pacific storm track confines oxidants in the boundary layer

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to a thin band along the cold front, as observed in previous studies (Kaneyasu et al., 2000; Sawa et al., 2007), with a significant proportion caught up in the warm sector ahead of it (e.g., Cooper et al., 2002). Continental boundary layer air with a greater abundance of European and North American oxidants is carried southeastwards in the cold outflow behind this front, but remains largely distinct from air of East Asian origin. The passage of this system over Japan is associated with a drop in midday temperature of 10 °C between 23 and 25 February and a pressure drop of 14 hPa followed by a rise of 30 hPa, see Fig. 13. The system first brings the footprint of Asian emissions to Tokyo in the warm sector, where Chinese CO reaches 80 ppb, and this is followed by 2–3 days of cold, dry continental outflow from higher latitudes with little influence from Chinese sources (Chinese CO below 10 ppb). European influence is higher in the post-frontal outflow, with European CO rising from 15 to 45 ppb over the period and a smaller rise for O_3 of 2.2 to 3.5 ppb. While there is some mixing of air influenced by European and East Asian sources in the post-frontal boundary layer outflow, as suggested by Liu et al. (2003), this episode demonstrates that the regional footprints can remain distinct within the timescales for transport from the continent to Japan.

The lifting of pollutants in the warm conveyor belt ahead of the cold front is clearly visible in the right panel of Fig. 13, where it brings Chinese contributions of more than 18 ppb of O_3 and 100 ppb of CO at 2–3 km altitude as it crosses Japan. At higher altitudes in the warm sector above 4 km the southwesterly flow brings air from southern and southeastern Asia, with O_3 reaching 12 ppb and CO reaching 180 ppb, and over Japan such high contributions from south Asian industrial and biomass burning sources are only seen under these conditions. The dry intrusion behind the cold front pushes this away and brings streamers of air down from the upper troposphere with 50–60 ppb of stratospheric O_3 which persist for about a day. At the surface, stratospheric contributions average 12 ppb in the air behind the front compared with about 6 ppb ahead of it on 23–24 February. The lowest levels of surface O_3 and CO over the week occur in the 24 h following passage of the cold front, when transport and photochemistry from local sources over Japan is suppressed and the contribution of

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continental sources remains low. The highest levels occur on 23 February in the 24 h before the system arrives, when surface O_3 exceeds 60 ppb for an 8-h period (for 5 h in the observations) and peaks at more than 70 ppb. Although early in the year, the fair-weather anticyclonic conditions contribute to substantial local O_3 production (20 ppb from Japanese sources) and the southwesterly flow brings as much as 14 ppb O_3 from Chinese sources. While this episode is not as severe as the major air quality exceedances later in spring shown in Table 6, it illustrates the transport mechanisms that allow substantial Chinese influence ahead of frontal systems where it may contribute to poor air quality over Japan. However, each synoptic system has a unique signature, and while this entrainment of continental air also occurred in the 20 March and 20 April exceedances leading to significant Chinese influence, in other cases the position of the Pacific High does not allow for this entrainment, and Chinese influence remains low, as on 12 April and 28 April. It would be valuable to explore the sensitivity of air quality exceedances in Japan to the position of these meteorological features in future work and to extend the period under consideration to account for the effects of interannual variability.

5 Conclusions

We have shown how local, regional and distant sources contribute to surface O_3 and CO over Japan in springtime and explored the transport pathways that lead to this influence. On a seasonal basis the largest contribution to daytime surface O_3 is from the stratosphere (20%), at a regional scale exceeding Japanese precursor sources (16%) which make a relatively small contribution in February but increase rapidly through the spring as photochemistry picks up. Local sources make relatively little net contribution to day-mean O_3 in this season due to strong O_3 removal at nighttime over polluted emission regions. Distant anthropogenic precursor emissions over China, Europe and North America make small but significant contributions to surface O_3 (7%, 6%, and 5% respectively), but together these sources make up 19% of O_3 over Japan, exceeding

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the local contribution and almost matching that from the stratosphere. The pattern for surface CO is similar, although Japanese sources make a smaller contribution (13%) and distant anthropogenic sources make a larger one (32%), with the largest effects from Europe (13%) during the spring season.

We have shown that these source contributions are highly variable during springtime and that clear meteorological signatures can be seen in both observed and modeled O_3 and CO at locations over the Western Pacific such as Minamitorishima. Spatial correlations with mean sea-level pressure and humidity indicate that on a climatological basis transport of pollutants from China to Japan occurs at low altitudes following the Pacific storm track, and is associated with the passage of cold fronts. Ozone contributions from Chinese sources typically reach 10–15 ppb during these episodes, which occur on average once or twice per week. In contrast, European influence is greatest in northwesterly flow representing continental boundary layer air caught up in the cold outflow some distance behind cold fronts. While O_3 and CO from Chinese and European sources are both transported to Japan at low levels behind these frontal systems, the influence from China is typically strongest in a band lying along the front, while that from Europe occurs later as cold continental air is entrained into the post-frontal outflow. Stratospheric contributions are largest above the capping inversions of anticyclones preceding (and following) these systems, but may reach as much as 20 ppb at the surface on occasions. However, examination of a number of case studies indicates that there is considerable variation in transport pathways between one synoptic meteorological system and the next that is partly driven by the timing and location of cyclogenesis and partly by the location of the Pacific High. We recommend that further studies focus on a more detailed meteorological classification of these systems so that long-range impacts on atmospheric composition can be characterized more clearly.

While meteorological boundaries such as fronts provide a clear separation of air from different nearby sources, the effects of more distant sources are more uniformly distributed, and their contributions remain significant during episodes of high O_3 over Tokyo in springtime. While the contribution from European and North American sources

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are typically close to their seasonal average during these episodes, contributions from China may often be substantially larger, although this is not always the case, depending on meteorological conditions. This study has focused on 2001, but we note that emissions of ozone precursors from Northern China are increasing due to rapid industrialization and increased vehicle ownership, and may already have increased by a factor of two since 2001 (e.g., Irie et al., 2005; Ohara et al., 2007). Our results suggest that this would account for an additional 2–4 ppb of daytime O_3 over Tokyo in spring, contributing to the observed trend. The magnitude of this O_3 contribution suggests that agencies regulating air quality in Japan should take account of long-range transport from the Asian mainland when examining exceedance of air quality standards in future.

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References

Akimoto, H.: Global air quality and pollution, Science, 302, 1716–1719, 2003. 10405

Akimoto, H., Mukai, H., Nishikawa, M., Murano, K., Hatakeyama, S., Liu, C.-M., Buhr, M., Hsu, K. J., Jaffe, D. A., Zhang, L., Honrath, R., Merrill, J. T., and Newell, R. E.: Long-range transport of ozone in the East Asian Pacific rim region, J. Geophys. Res., 101, 1999–2010, 1996.

Berntsen, T. K., Isaksen, I. S. A., Wang, W.-C., and Liang, X. Z.: Impacts of increased anthropogenic emissions in Asia on tropospheric ozone and climate – a global 3-D model study, Tellus, 48, 13–32, 1996. 10405, 10410

Berntsen, T. K., Karlsdottir, S., and Jaffe, D. A.: Influence of Asian emissions on the composition of air reaching the Northwestern United States, Geophys. Res. Lett., 26, 2171–2174, 1999. 10405

Bey, I., Jacob, D. J., Logan, J. A., and Yantosca, R. M.: Asian chemical outflow to the Pacific: origins, pathways and budgets, J. Geophys. Res., 106, 23097–23114, 2001. 10406

Carmichael, G. R., Uno, I., Phadnes, M., Zhang, Y., and Sunwoo, Y.: Tropospheric ozone

ACPD

11, 10403–10448, 2011

production and transport in the springtime in east Asia, J. Geophys. Res., 103, 10649–10671, 1998. 10406, 10416

Carmichael, G. R., Tang, Y., Kurata, G., Uno, I., Streets, D. G., Thongboonchoo, N., Woo, J.-H., Guttikunda, S., White, A., Wang, T., Blake, D. R., Atlas, E., Fried, A., Potter, B., Avery, M. A., Sachse, G. W., Sandholm, S. T., Kondo, Y., Talbot, R. W., Bandy, A., Thorton, D., and Clarke, A. D.: Evaluating regional emission estimates using the TRACE-P observations, J. Geophys. Res., 108(D21), 8810, doi:10.1029/2002JD003116, 2003.

Cooper, O. R., Moody, J. L., Parrish, D. D., Trainer, M., Ryerson, T. B., Holloway, J. S., Hübler, G., Fehsenfeld, F. C., and Evans, M. J.: Trace gas composition of midlatitude cyclones over the western North Atlantic Ocean: A conceptual model, J. Geophys. Res., 107(D7), 4056, doi:10.1029/2001JD000901, 2002. 10416, 10419

Dentener, F., Stevenson, D., Ellingsen, K., van Noije, T., Schultz, M., Amann, M., Atherton, C., Bell, N., Bergmann, D., Bey, I., Bouwman, L., Butler, T., Cofala, J., Collins, B., Drevet, J., Doherty, R., Eickhout, B., Eskes, H., Fiore, A., Gauss, M., Hauglustaine, D., Horowitz, L., Isaksen, I. S. A., Josse, B., Lawrence, M., Krol, M., Lamarque, J. F., Montanaro, V., Müller, J. F., Pauch, V. H., Pitari, G., Pyle, J., Rast, S., Rodriguez, J., Sanderson, M., Savage, N. H., Shindell, D., Strahan, S., Szopa, S., Sudo, K., Van Dingenen, R., Wild, O., and Zeng, G.: The global atmospheric environment for the next generation, Environ. Sci. Technol., 40, 3586–3594, 2006. 10405

Duncan, B. N., Martin, R. V., Staudt, A. C., Yevich, R., and Logan, J. A.: Interannual and seasonal variability of biomass burning emissions constrained by satellite observations, J. Geophys. Res., 108(D9), 4040, doi:10.1029/2002JD002378, 2003. 10411

Fiore, A. M., Jacob, D. J., Bey, I., Yantosca, R. M., Field, B. D., Fusco, A. C., and Wilkinson, J. G.: Background ozone over the United States in summer: Origin, trend, and contribution to pollution episodes, J. Geophys. Res., 107(D15), 4275, doi:10.1029/2001JD000982, 2002. 10405, 10414

Fiore, A. M., Jacob, D. J., Liu, H., Yantosca, R. M., Fairlie, T. D., and Li, Q. B.: Variability in surface ozone background over the United States: Implications for air quality policy, J. Geophys. Res., 108, 4787, doi:10.1029/2003JD003855, 2003. 10405

Fiore, A. M., Dentener, F. J., Wild, O., Cuvelier, C., Schultz, M. G., Hess, P., Textor, C., Schulz, M., Doherty, R. M., Horowitz, L. W., MacKenzie, I. A., Sanderson, M. G., Shindell, D. T., Stevenson, D. S., Szopa, S., Van Dingenen, R., Zeng, G., Atherton, C., Bergmann, D., Bey, I., Carmichael, G., Collins, W. J., Duncan, B. N., Faluvegi, G., Folberth, G., Gauss, M., Gong,

ACPD
11, 10403–10448, 2011

S., Hauglustaine, D., Holloway, T., Isaksen, I. S. A., Jacob, D. J., Jonson, J. E., Kaminski, J. W., Keating, T. J., Lupu, A., Marmer, E., Montanaro, V., Park, R. J., Pitari, G., Pringle, K. J., Pyle, J. A., Schroeder, S., Vivanco, M. G., Wind, P., Wojcik, G., Wu, S., and Zuber, A.: Multi-model estimates of intercontinental source-receptor relationships for ozone pollution, J. Geophys. Res., 114, D04301, doi:10.1029/2008JD010816, 2009. 10405

Fuelberg, H. E., Kiley, C. M., Hannan, J. R., Westberg, D. J., Avery, M. A., and Newell, R. E.: Meteorological conditions and transport pathways during the Transport and Chemical Evolution over the Pacific (TRACE-P) experiment, J. Geophys. Res., 108(D20), 8782, doi:10.1029/2002JD003092, 2003. 10416

Heald, C. L., Jacob, D. J., Fiore, A. M., Emmons, L. K., Gille, J. C., Deeter, M. N., Warner, J., Edwards, D. P., Crawford, J. H., Hamlin, A. J., Sachse, G. W., Browell, E. V., Avery, M. A., Vay, S. A., Westberg, D. J., Blake, D. R., Singh, H. B., Sandholm, S. T., Talbot, R. W., and Fuelberg, H. E.: Asian outflow and transpacific transport of carbon monoxide and ozone pollution: An integrated satellite, aircraft and model perspective, J. Geophys. Res., 108(D24), 4804, doi:10.1029/2003JD003507, 2003. 10405, 10406

Holloway, T., Fiore, A., and Galanter Hastings, M.: Intercontinental Transport of Air Pollution: Will emerging science lead to a new hemispheric treaty?, Environ. Sci. Technol., 37, 4535–4542, 2003. 10405

Holloway, T., Sakurai, T., Han, Z., Ehlers, S., Spak, S. N., Horowitz, L. W., Carmichael, G. R., Streets, D. G., Hozumi, Y., Ueda, H., Park, S. U., Fung, C., Kajino, M., Thongboonchoo, N., Engardt, M., Bennet, C., Hayami, H., Sartelet, K., Wang, Z., Matsuda, K., and Amann, M.: MICS-Asia II: Impact of global emissions on regional air quality in Asia, Atmos. Environ., 42, 3543–3561, 2008.

Irie, H., Sudo, K., Akimoto, H., Richter, A., Burrows, J. P., Wagner, T., Wenig, M., Beirle, S., Kondo, Y., Sinyakov, V. P., and Goutail, F.: Evaluation of long-term tropospheric NO_2 data obtained by GOME over East Asia in 1996–2002, Geophys. Res. Lett., 32, L11810, doi:10.1029/2005GL022770, 2005. 10406, 10422

Jacob, D. J., Logan, J. A., and Murti, P. P.: Effect of rising Asian emissions on surface ozone in the United States, Geophys. Res. Lett., 26, 2175–2178, 1999. 10405

Jacob, D. J., Crawford, J. H., Kleb, M. M., Connors, V. S., Bendura, R. J., Raper, J. L., Sachse, G. W., Gille, J. C., Emmons, L., and Heald, C. L.: Transport and Chemical Evolution over the Pacific (TRACE-P) aircraft mission: Design, execution and first results, J. Geophys. Res., 108(D20), 9000, doi:10.1029/2002JD003276, 2003. 10407

ACPD
11, 10403–10448, 2011

Jaeglé, L., Jaffe, D. A., Price, H. U., Weiss-Penzias, P., Palmer, P. I., Evans, M. J., Jacob, D. J., and Bey, I.: Sources and budgets for CO and O_3 in the northeastern Pacific during the spring of 2001: Results from the PHOBEA-II Experiment, J. Geophys. Res., 108(D20), 8802, doi:10.1029/2002JD003121, 2003. 10406

Jaffe, D. A., McKendry, I., Anderson, T., and Price, H.: Six "new" episodes of trans-Pacific transport of air pollutants, Atmos. Environ., 37(3), 391–404, 2003. 10405, 10406

Kajii, Y., Someno, K., Tanimoto, H., Hirokawa, J., Akimoto, H., Katsuno, T., and Kawara, J.: Evidence for the seasonal variation of photochemical activity of tropospheric ozone: Continuous observation of ozone and CO at Happo, Japan, Geophys. Res. Lett., 25, 3505–3508, 1998. 10413

Kaneyasu, N., Takeuchi, K., Fujita, S., Uno, I., and Sasaki, H.: Outflow patterns of pollutants from east Asia to the North Pacific in the winter monsoon, J. Geophys. Res., 105, 17361–17377, 2000. 10406, 10416, 10419

Lee, S.-H., Akimoto, H., Nakane, H., Kurnosenko, S., and Kinjo, Y.: Lower tropospheric ozone trend observed in 1989–1997 at Okinawa, Japan, Geophys. Res. Lett., 25(10), 1637–1640, 1998. 10405

Liang, Q., Jaegle, L., Jaffe, D. A., Weiss-Penzias, P., Heckman, A., and Snow, J. A.: Long-range Transport to the Northeast Pacific: Seasonal variations and transport pathways of carbon monoxide, J. Geophys. Res., 109(D23), D23S07, doi:10.1029/2003JD004402, 2004. 10406, 10413

Liang, Q., Jaegle, L., and Wallace, J. M.: Meteorological indices for Asian outflow and transpacific transport on daily to interannual timescales, J. Geophys. Res., 110, D18308, doi:10.1029/2005JD005788, 2005. 10418

Lin, C.-Y. C., Jacob, D. J., and Fiore, A. M.: Trends in exceedances of the ozone air quality standard in the continental United States, 1980–1998, Atmos. Environ., 35, 3217–3228, 2001. 10405

Lin, M., Holloway, T., Carmichael, G. R., and Fiore, A. M.: Quantifying pollution inflow and outflow over East Asia in spring with regional and global models, Atmos. Chem. Phys., 10, 4221–4239, doi:10.5194/acp-10-4221-2010, 2010. 10406

Lippmann, M.: Health effects of tropospheric ozone, Environ. Sci. Technol., 25, 1954–1962, 1991. 10405

Liu, H., Bey, I., Yantosca, R. M., Duncan, B. N., and Sachse, G. W.: Transport pathways for Asian combustion outflow over the Pacific: Interannual and seasonal variations, J. Geophys.

ACPD
11, 10403–10448, 2011

Res., 108, 8786, doi:10.1029/2002JD003102, 2003. 10406, 10409, 10416, 10417, 10419
Mauzerall, D. L. and Wang, X.: Protecting Agricultural Crops from the Effects of Tropospheric Ozone Exposure: Reconciling Science and Standard Setting in the United States, Europe and Asia, Annu. Rev. Energ. Env., 26, 237–268, 2001. 10405
Mauzerall, D. L., Narita, D., Akimoto, H., Horowitz, L., Walters, S., Hauglustaine, D. A., and Brasseur, G.: Seasonal Characteristics of Tropospheric Ozone Production and Mixing Ratios Over East Asia: A Global Three-dimensional Chemical Transport Model Analysis, J. Geophys. Res., 105, 17895–17910, 2000.
Mickley, L. J., Murti, P. P., Jacob, D. J., Logan, J. A., Rind, D., and Koch, D.: Radiative forcing from tropospheric ozone calculated with a unified chemistry-climate model, J. Geophys. Res., 104, 30153–30172, 1999. 10405
Miyazaki, Y., Kondo, Y., Koike, M., Fuelberg, H. E., Kiley, C. M., Kita, K., Takegawa, N., Sachse, G. W., Flocke, F., Weinheimer, A. J., Singh, H. B., Eisele, F. L., Zondlo, M., Talbot, R. W., Sandholme, S. T., Avery, M. A., and Blake, D. R.: Synoptic-scale transport of reactive nitrogen over the western Pacific in spring, J. Geophys. Res., 108(D20), 8788, doi:10.1029/2002JD003248, 2003. 10406, 10416
Nagashima, T., Ohara, T., Sudo, K., and Akimoto, H.: The relative importance of various source regions on East Asian surface ozone, Atmos. Chem. Phys., 10, 11305–11322, doi:10.5194/acp-10-11305-2010, 2010. 10414
Naja, M. and Akimoto, H.: Contribution of regional and long-range transport to the Asia-Pacific region: Analysis of long-term ozonesonde data over Japan, J. Geophys. Res., 109, D21306, doi:10.1029/2004JD004687, 2004. 10405
Network Center for EANET: EANET Data on Acid Deposition in the East Asia Region, Acid Deposition and Oxidant Research Center (ADORC), Niigata, Japan, available at: http://www.eanet.cc/, 2005. 10408
Ohara, T. and Sakata, T.: Long-term variation of photochemical oxidants over Japan, J. Japan Soc. Atmos. Environ., 38, 47–54, 2003 (in Japanese). 10405
Ohara, T., Akimoto, H., Kurokawa, J., Horii, N., Yamaji, K., Yan, X., and Hayasaka, T.: An Asian emission inventory of anthropogenic emission sources for the period 1980–2020, Atmos. Chem. Phys., 7, 4419–4444, doi:10.5194/acp-7-4419-2007, 2007. 10406, 10422
Olivier, J. G. J., Bouwman, A. F., van der Maas, C. W. M., Berdowski, J. J. M., Veldt, C., Bloos, J. P. J., Visschedijk, A. J. H., Zandveld, P. Y. J., and Haverlag, J. L.: Description of EDGAR Version 2.0, RIVM/TNO report 771060 002, RIVM, Bilthoven, The Netherlands, 1996. 10408,

ACPD
11, 10403–10448, 2011

10440
Parrish, D. D., Holloway, J. S., Trainer, M., Murphy, P. C., Fehsenfeld, F. C., and Forbes, G. L.: Export of North American ozone pollution to the North Atlantic Ocean, Science, 259, 1436–1439, 1993. 10413
Pochanart, P., Hirokawa, J., Kajii, Y., Akimoto, H., and Nakao, M.: Influence of regional-scale anthropogenic activity in northeast Asia on seasonal variations of surface ozone and carbon monoxide observed at Oki, Japan, J. Geophys. Res., 104(D20), 3621–3361, 1999. 10413
Pochanart, P., Akimoto, H., Maksyutov, S., and Staehelin, J.: Surface ozone at the Swiss Alpine site Arosa: the hemispheric background and the influence of large-scale anthropogenic emissions, Atmos. Environ. 35, 5553–5566, 2001. 10405
Pochanart, P., Akimoto, H., Kinjo, Y., and Tanimoto, H.: Surface ozone at four remote island sites and the preliminary assessment of exceedances of its critical level in Japan, Atmos. Environ., 36, 4235–4250, 2002. 10405
Prather, M. and Ehhalt, D.: Atmospheric Chemistry and Greenhouse Gases, Climate Change 2001, edited by: Houghton, J. T., Ding, Y., and Griggs, D. J., 349–416, Cambridge Univ. Press, Cambridge, 2001.
Prather, M., Gauss, M., Berntsen, T., Isaksen, I., Sundet, J., Bey, I., Brasseur, G., Dentener, F., Derwent, R., Stevenson, D., Grenfell, L., Hauglustaine, D., Horowitz, L., Jacob, D., Mickley, L., Lawrence, M., von Kuhlmann, R., Muller, J.-F., Pitari, G., Rogers, H., Johnson, M., Pyle, J., Law, K., van Weele, M., and Wild, O.: Fresh air in the 21st century?, Geophys. Res. Lett., 30(2), 1100, doi:10.1029/2002GL016285, 2003. 10405
Roelofs, G. J. and Lelieveld, J.: Model study of the influence of cross-tropopause ozone transports on tropospheric ozone levels, Tellus, Ser. B, 49, 38–55, 1997. 10412
Sawa, Y., Tanimoto, H., Yonemura, S., Matsueda, H., Wada, A., Taguchi, S., Hayasaka, T., Tsuruta, H., Tohjima, Y., Mukai, H., Kikuchi, N., Katagiri, S., and Tsuboi, K.: Widespread pollution events of carbon monoxide observed over the western North Pacific during the East Asian Regional Experiment (EAREX) 2005 campaign, J. Geophys. Res., 112, D22S26, doi:10.1029/2006JD008055, 2007. 10406, 10409, 10419
Stohl, A.: A 1-year Lagrangian "climatology" of airstreams in the Northern Hemisphere troposphere and lowermost stratosphere, J. Geophys. Res., 106, 7263–7279, 2001. 10406
Streets, D. G., Yarber, K. F., Woo, J.-H., and Carmichael, G. R.: Biomass Burning in Asia: Annual and seasonal estimates and atmospheric emissions, Global Biogeochem. Cycles, 17(4), 1099, doi:10.1029/2003GB002040, 2003. 10408, 10411, 10440

ACPD
11, 10403–10448, 2011

Tanimoto, H., Sawa, Y., Matsueda, H., Uno, I., Ohara, T., Yamaji, K., Kurokawa, J., and Yonemura, S.: Significant latitudinal gradient in the surface ozone spring maximum over East Asia, Geophys. Res. Lett., 32, L21805, doi:10.1029/2005GL023514, 2005. 10406

Task Force on Hemispheric Transport of Air Pollutants (TF-HTAP): Hemispheric Transport of Air Pollution 2007, Air Pollut. Stud. 16, edited by: Keating, T. J. and Zuber, A., UNECE, Geneva, Switzerland, available at: http://www.htap.org/, 2007. 10405

Thompson, A. M.: The oxidising capacity of the earth's atmosphere: Probable past and future changes, Science, 256, 1157–1165, 1992. 10405

Tokyo Metropolitan Government: Report of the Committee for Photochemical Oxidant and its Control, Tokyo Metropolitan Government, Tokyo, Japan, 2005. 10406

Tsutsumi, Y., Mori, K., Ikegami, M., Tashiro, T., and Tsuboi, K.: Long-term trends of greenhouse gases in regional and background events observed during 1998–2004 at Yonagunijima located to the east of the Asian continent, Atmos. Environ., 40, 5868–5879, 2006. 10408

Uno, I., Jang, E.-S., Murano, K., Shimohara, T., Oishi, O., Utsunomiya, A., Hatakeyama, S., Tang, X., and Kim, Y.-P.: Wintertime intermittent transboundary air pollution over East Asia simulated by a long-range transport model, Global Environmental Research, 4, 3–12, 2000.

Wang, X. and Mauzerall, D. L.: Characterizing Distributions of Surface Ozone and its Impact on Grain Production in China, Japan and South Korea: 1990 and 2020, Atmos. Environ., 38, 4383–4402, 2004. 10405

Wild, O. and Akimoto, H.: Intercontinental transport of ozone and its precursors in a 3-D global model, J. Geophys. Res., 106, 27729–27744, 2001. 10405, 10410

Wild, O. and Prather, M. J.: Excitation of the primary tropospheric chemical mode in a global 3-D model, J. Geophys. Res., 105, 24647–24660, 2000. 10407

Wild, O. and Prather, M. J.: Global tropospheric ozone modeling: Quantifying errors due to grid resolution, J. Geophys. Res., 111, D11305, doi:10.1029/2005JD006605, 2006. 10410

Wild, O., Sundet, J. K., Prather, M. J., Isaksen, I. S. A., Akimoto, H., Browell, E. V., and Oltmans, S. J.: CTM Ozone Simulations for Spring 2001 over the Western Pacific: Comparisons with TRACE-P lidar, ozonesondes and TOMS columns, J. Geophys. Res., 108, 8826, doi:10.1029/2002JD003283, 2003. 10406, 10407, 10408

Wild, O., Pochanart, P., and Akimoto, H.: Trans-Eurasian transport of ozone and its precursors, J. Geophys. Res., 109, D11302, doi:10.1029/2003JD004501, 2004. 10413, 10417

Wilkening, K. E., Barrie, L., and Engle, M.: Trans-Pacific air pollution, Science, 290, 65–67, 2000. 10405

ACPD
11, 10403–10448, 2011

Wilkening, K. E.: Trans-Pacific air pollution: scientific evidence and political implication, Water Air Soil Pollut., 130(1–4), 1825–1830, 2001. 10405

Wu, S., Duncan, B. N., Jacob, D. J., Fiore, A. M., and Wild, O.: Chemical nonlinearities in relating intercontinental ozone pollution to anthropogenic emissions, Geophys. Res. Lett., 36, L05806, doi:10.1029/2008GL036607, 2009. 10412

Yamaji, K., Ohara, T., Uno, I., Tanimoto, H., Kurokawa, J., and Akimoto, H.: Analysis of the seasonal variation of ozone in the boundary layer in east Asia using the Community Multi-scale Air Quality model: What controls surface ozone levels over Japan?, Atmos. Environ., 40(10), 1856–1868, doi:10.1016/j.atmosenv.2005.10.067, 2006. 10406

Yienger, J. J., Klonecki, A. A., Levy II, H., Moxim, W. J., and Carmichael, G. R.: An evaluation of chemistry's role in the winter-spring ozone maximum found in the northern midlatitude free troposphere, J. Geophys. Res., 104, 3655–3667, 1999.

Yienger, J. J., Galanter, M., Holloway, T. A., Phadnis, M. J., Guttikunda, S. K., Carmichael, G. R., Moxim, W. J., and Levy II, H.: The episodic nature of air pollution transport from Asia to North America, J. Geophys. Res., 105, 26931–26945, 2000. 10405, 10416

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Table 1. List of EANET Measurement Sites and WMO/GAW Stations Used.

Site/Station	Organization	Latitude	Longitude	Altitude	Characteristics
Rishiri	EANET	45°07′ N	141°12′ E	40 m	Remote Island
Tappi	EANET	41°15′ N	140°21′ E	105 m	Remote
Sadoseki	EANET	38°14′ N	138°24′ E	110 m	Rural
Happo	EANET	36°42′ N	137°48′ E	1850 m	Remote Mountain
Oki	EANET	36°17′ N	133°11′ E	90 m	Remote Island
Ijira	EANET	35°34′ N	136°41′ E	140 m	Urban
Banryu	EANET	34°41′ N	131°48′ E	60 m	Urban
Yusuhara	EANET	33°22′ N	132°56′ E	225 m	Rural
Ogasawara	EANET	27°05′ N	142°13′ E	230 m	Remote Island
Hedo	EANET	26°52′ N	128°15′ E	50 m	Remote
Ryori	WMO/GAW	39°02′ N	141°49′ E	230 m	Rural
Tsukuba	WMO/GAW	36°03′ N	140°08′ E	25 m	Urban
Yonagunijima	WMO/GAW	24°28′ N	123°01′ E	30 m	Remote Island
Minamitorishima	WMO/GAW	24°18′ N	153°58′ E	8 m	Remote Island

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Table 2. Statistical Summary of Observed and Modeled Hourly Surface O_3 (in ppb) in February–April 2001.

Site/Station	*n*	Mean O_3			Mean Bias		RMSE	
		Obs.	T21	T63	T21	T63	T21	T63
				Remote				
Rishiri	1175	44.1	45.8	44.3	1.8	0.2	7.3	9.2
Tappi	2013	55.0	54.7	49.8	−0.3	−5.2	8.0	10.3
Happo	2087	62.8	62.1	58.7	−0.6	−4.1	9.8	9.1
Oki	1730	50.2	59.4	55.8	9.3	5.6	12.5	9.2
Ogasawara	2039	40.0	41.4	48.6	1.5	8.6	16.2	13.4
Hedo	2103	53.9	51.8	54.8	−2.1	0.9	11.9	7.1
Yonagunijima	2128	48.6	57.3	53.5	8.7	4.9	13.7	9.7
Minamitorishima	2096	35.7	35.5	37.7	−0.2	2.0	8.1	6.1
				Rural[a]				
Ryori	421	44.9	55.4	54.4	10.5	9.5	13.2	12.0
Sadoseki	445	48.8	59.5	52.8	10.8	4.0	17.8	13.6
Yusuhara	445	35.1	60.7	57.6	25.6	22.6	27.8	24.7
				Urban[a]				
Tsukuba	445	51.8	63.2	60.6	11.4	8.8	18.0	15.8
Ijira	445	52.9	65.3	60.7	12.4	7.8	17.8	14.7
Banryu	445	53.5	66.7	61.0	13.2	7.4	16.1	12.0

[a] Only afternoon data (12:00–16:00 JST) used at rural/urban sites.

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Table 3. Statistical Summary of Observed and Modeled Hourly Surface CO (in ppb) in February–April 2001.

Site/Station	*n*	Mean CO			Mean Bias		RMSE	
		Obs.	T21	T63	T21	T63	T21	T63
Ryori	1020	228.8	239.2	212.7	10.3	−16.1	41.7	42.5
Yonagunijima	1991	222.9	219.2	206.5	−3.7	−16.4	66.2	54.8
Minamitorishima	1896	149.7	140.1	144.6	−9.6	−5.1	23.2	18.0

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Table 4. Source Contributions to Surface CO in Tokyo During Spring 2001.

	Feb ppb (%)		Mar ppb (%)		Apr ppb (%)		Feb–Apr ppb (%)	
JP	26.8	(12.1)	24.8	(10.7)	34.1	(15.4)	28.5	(12.7)
CH	24.3	(11.0)	25.3	(11.0)	20.7	(9.4)	23.4	(10.4)
EU	30.3	(13.7)	30.6	(13.3)	26.7	(12.1)	29.3	(13.0)
NA	20.0	(9.0)	20.0	(8.7)	17.6	(7.9)	19.2	(8.5)
SA	4.0	(1.8)	4.4	(1.9)	4.1	(1.8)	4.2	(1.9)
BB	2.5	(1.1)	4.8	(2.1)	6.0	(2.7)	4.5	(2.0)
Total CO	221.2		231.0		221.7		224.8	

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Table 5. Source Contributions to Afternoon Surface O_3 in Tokyo During Spring 2001.

	Feb ppb (%)		Mar ppb (%)		Apr ppb (%)		Feb–Apr ppb (%)	
JP	5.6	(10.6)	8.2	(13.3)	14.7	(21.7)	9.4	(15.6)
CH	4.2	(8.0)	4.6	(7.4)	4.5	(6.6)	4.4	(7.3)
EU	2.9	(5.4)	4.2	(6.9)	4.5	(6.6)	3.9	(6.4)
NA	3.1	(5.9)	3.1	(5.1)	3.0	(4.4)	3.0	(5.0)
SA	0.6	(1.2)	0.6	(0.9)	0.4	(0.6)	0.5	(0.9)
BB	0.2	(0.3)	0.3	(0.5)	0.4	(0.6)	0.3	(0.5)
Strat	11.8	(22.4)	13.2	(21.5)	11.9	(17.5)	12.2	(20.2)
Total O_3	52.6		61.2		68.1		60.5	

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Table 6. Source Contributions to Surface O_3 (ppb) in Tokyo During High O_3 Episodes (O_3>80 ppb) in Spring 2001.

Date	Duration	Mean O_3	JP	CH	EU	NA	SA	BB	Strat
20 March	1 h	80.1	18.5	14.5	4.5	3.0	0.4	0.4	10.4
6 April	5 h	82.1	26.6	5.6	4.3	2.8	0.4	0.4	12.0
12 April	5 h	83.8	42.4	2.2	2.6	2.0	0.4	0.5	8.1
17 April	8 h	86.3	24.8	11.1	4.8	3.1	0.4	0.4	9.2
20 April	4 h	81.4	21.3	14.8	3.2	2.5	0.3	0.4	7.6
28 April	2 h	80.8	27.4	3.1	4.6	2.7	0.4	0.4	9.9
Contrasting episodes with monthly mean									
Episode Mean		83.5	28.1	8.3	4.0	2.7	0.4	0.4	9.4
April Mean		68.1	14.7	4.5	4.5	3.0	0.4	0.4	11.9

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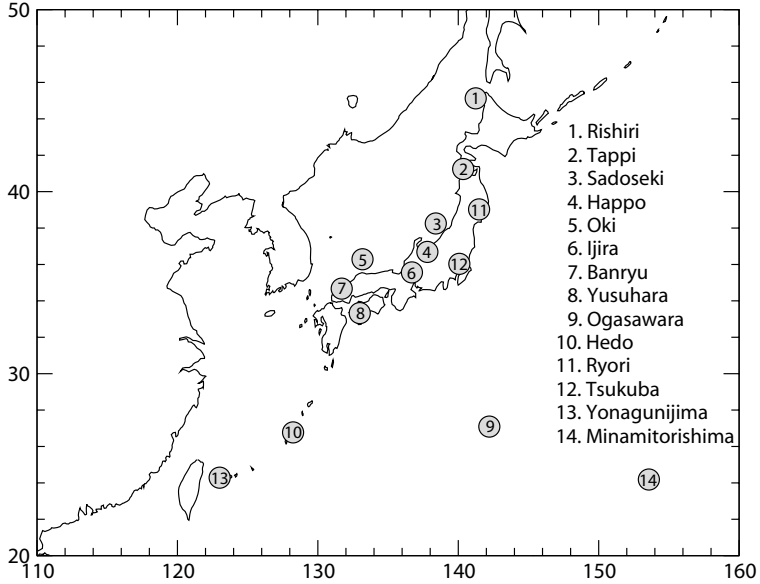

Fig. 1. Locations of EANET and WMO/GAW stations used in evaluating model performance.

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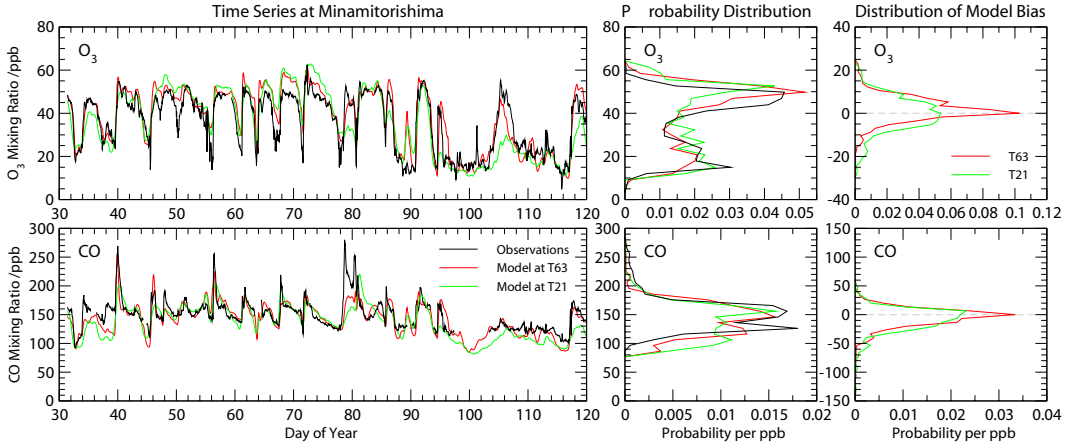

Fig. 2. Timeseries of hourly-mean surface O_3 and CO observations at Minamitorishima WMO/GAW station in February–April 2001 and FRSGC/UCI CTM results at T21 and T63 resolution. Right-hand panels show the probability distribution over the period and the distribution of the hourly model bias at each resolution.

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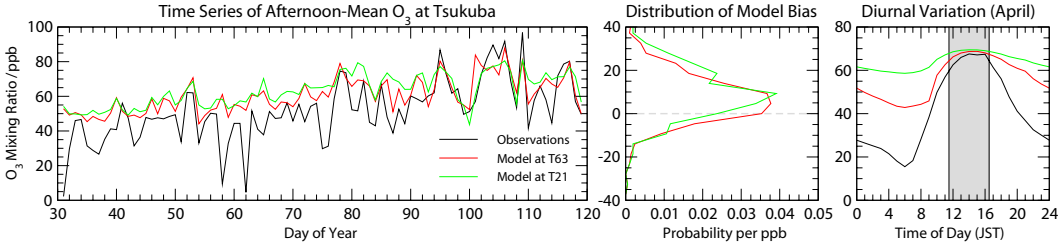

Fig. 3. Comparison of afternoon-mean O_3 observations (12:00–16:00 h JST) at Tsukuba WMO/GAW station (black) with CTM results at T21 (green) and T63 (red) resolution. Right-hand panels show the distribution of the hourly model bias and the monthly mean diurnal variation in observed and modelled O_3 in April.

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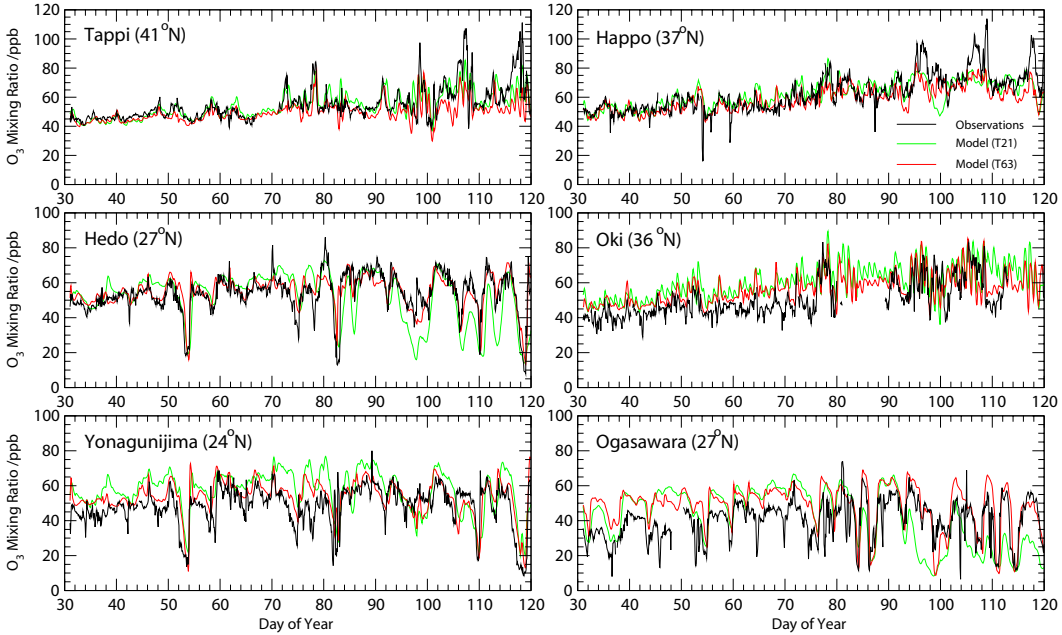

Fig. 4. Time series of hourly-mean O_3 observations at selected remote EANET and WMO/GAW measurement sites in February–April 2001 (black) and FRSGC/UCI CTM results at T21 (green) and T63 (red) resolution.

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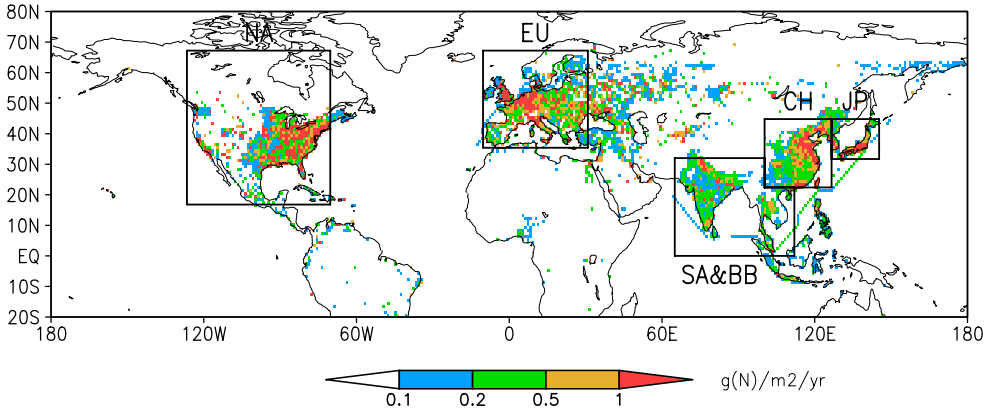

Fig. 5. Distribution of anthropogenic surface NO_x emissions ($g\,N\,m^{-2}\,yr^{-1}$) used in this study (Olivier et al., 1996; Streets et al., 2003) showing the principal source regions considered here.

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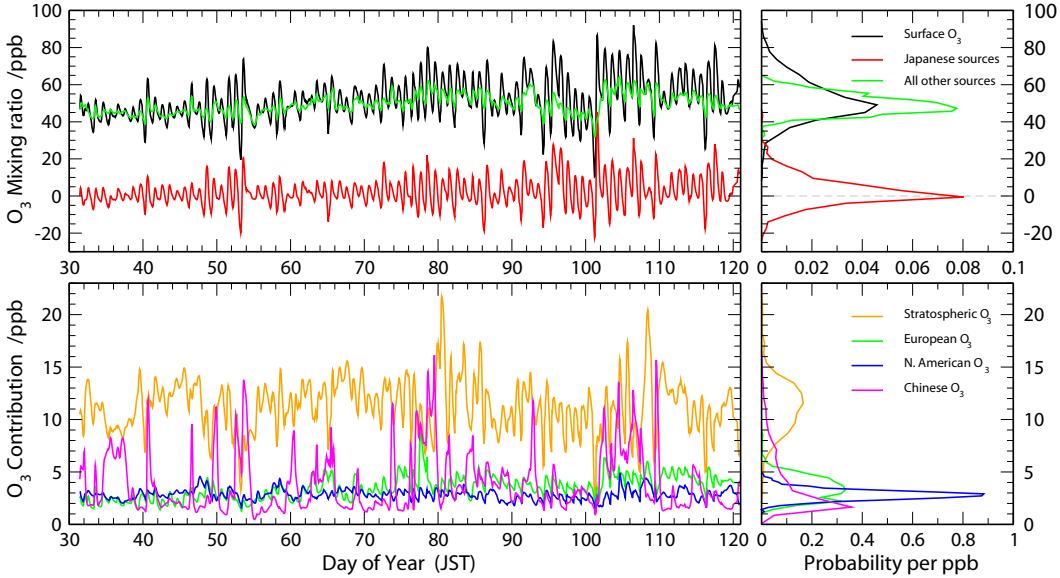

Fig. 6. Hourly mean surface O_3 mixing ratios in Tokyo in February–April 2001 from the FRSGC/UCI CTM showing **(a)** total surface O_3 (black), O_3 from Japanese sources (red) and background O_3 from other sources (green), and **(b)** contributions to O_3 from Chinese (magenta), European (green) and North American (blue) sources and from the stratosphere (orange). Right-hand panels show the probability distribution over the three-month period.

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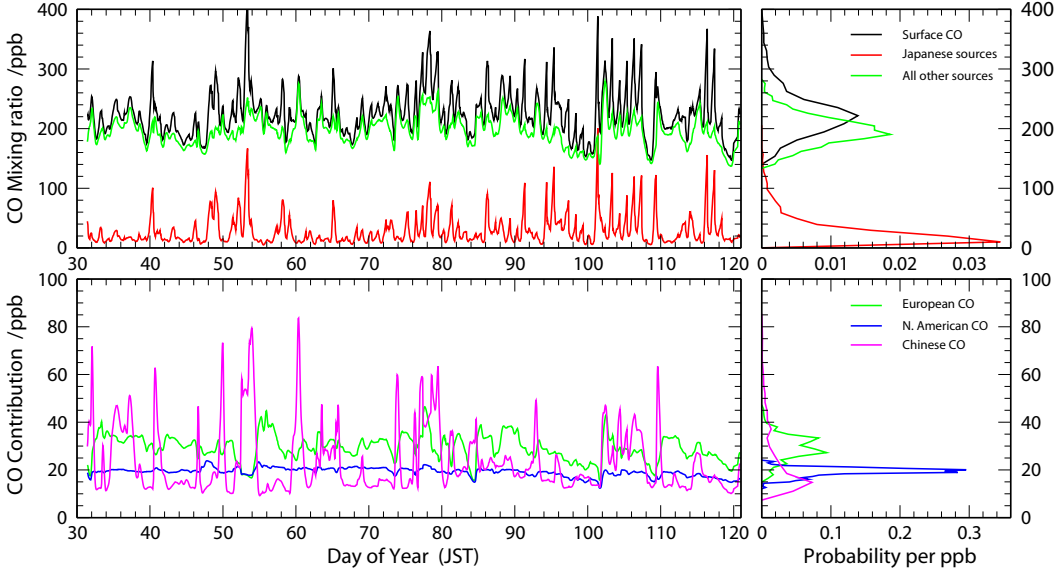

Fig. 7. Hourly mean surface CO mixing ratios in Tokyo in February–April 2001 from the FRSGC/UCI CTM showing **(a)** total surface CO (black), CO from Japanese sources (red) and background CO from other sources (green), and **(b)** contributions to CO from Chinese (magenta), European (green), and North American (blue) sources. Right-hand panels show the probability distribution over the three-month period.

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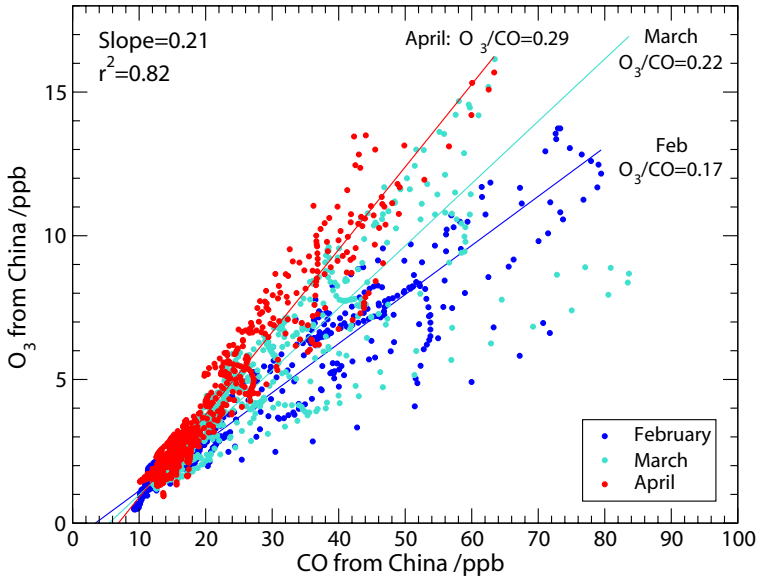

Fig. 8. Relationship between Chinese contributions to surface O_3 and CO over Tokyo revealing the increase in photochemical activity between February and April in 2001.

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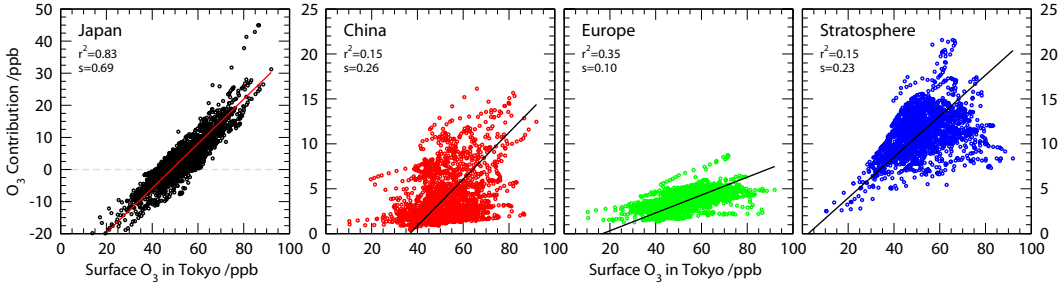

Fig. 9. Contributions of Japanese, Chinese and European industrial sources and transport from the stratosphere to surface O_3 over Tokyo as a function of total surface O_3. Graphs are labelled with the correlation coefficient (r) and slope (s).

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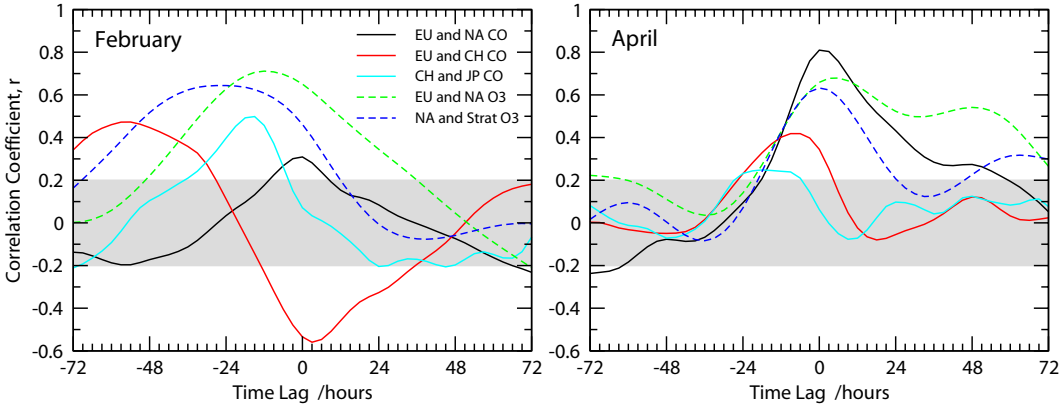

Fig. 10. Correlation between the contributions of key source regions to O_3 and CO over Tokyo in February (left) and April (right).

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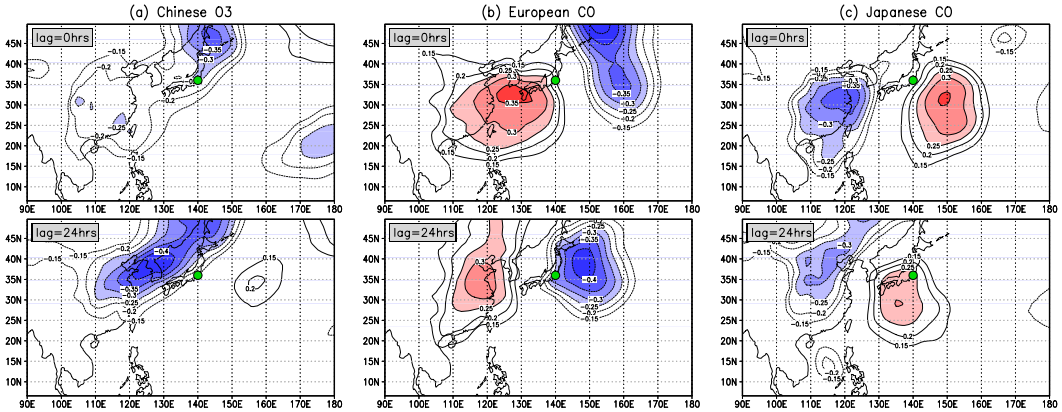

Fig. 11. Correlation between mean sea-level pressure and the contributions of **(a)** Chinese O_3, **(b)** European CO and **(c)** Japanese CO over Tokyo (marked with a circle) in spring 2001. Positive correlations are highlighted in red and negative correlations in blue. Lower panels show the correlation coefficient lagged by 24 h, and thus the relationship with mean sea-level pressure one day earlier.

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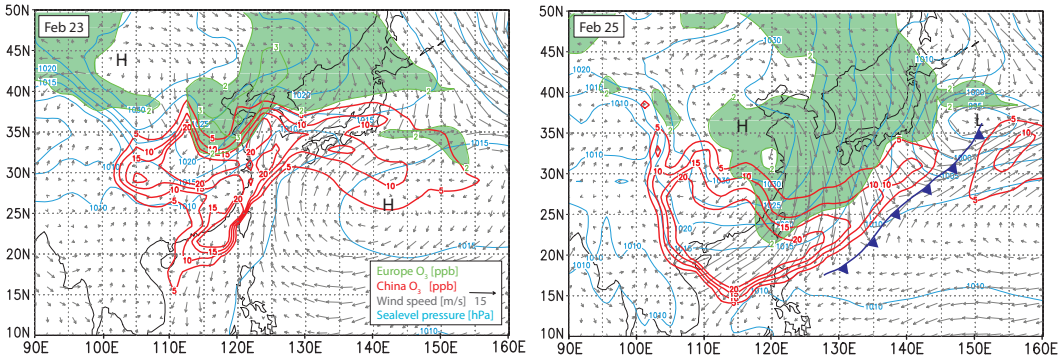

Fig. 12. Chinese and European contributions to O_3 and CO over Japan at 00:00 UTC on 23 and 25 February 2001 showing the development and movement of air masses associated with a developing frontal system (approximate position on 25 February marked in blue).

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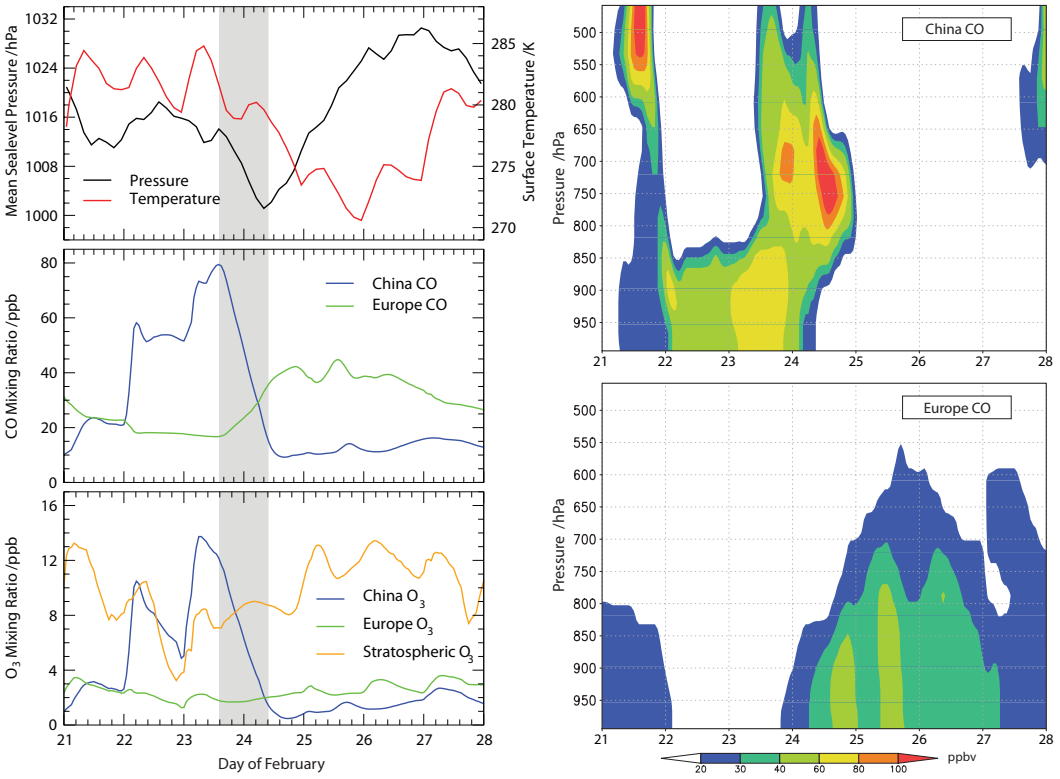

Fig. 13. Variations in surface pressure and temperature over Tokyo during the week of 21–28 February 2001, showing the passage of the frontal system shown in Fig. 12 (highlighted in grey) and the associated changes in the contribution of distant sources to O_3 and CO (left panels). Vertical profiles of the contributions of China and Europe to CO over Tokyo between the surface and 500 hPa (about 5 km) are shown in the righthand panels, highlighting the passage of the cold front on 24 February and elevated levels of CO from China aloft.

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