

Contributions of regional and intercontinental transport to surface ozone in the Tokyo area

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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 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



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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; Tanimoto et al., 2009). This is of particular concern in the metropolitan area of Tokyo, 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.

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 have on springtime air quality in Japan.

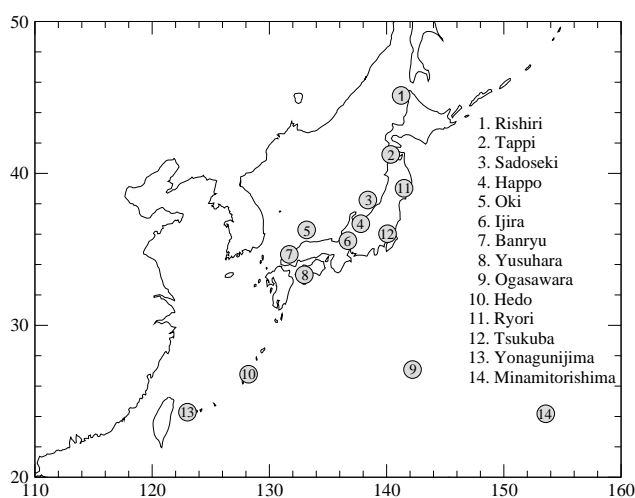
2 Model and evaluation against observations

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). 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 suburban

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	Suburban
Banryu	EANET	34°41' N	131°48' E	60 m	Suburban
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	Suburban
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

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

based on their chemical environment. Eight clean coastal or mountain sites are categorized as remote, three sites showing strong pollution signatures are designated as suburban, and the remaining three sites showing some influence from local sources are denoted as rural. These observations are compared with hourly model results sampled from the nearest grid box at each resolution.

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 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 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

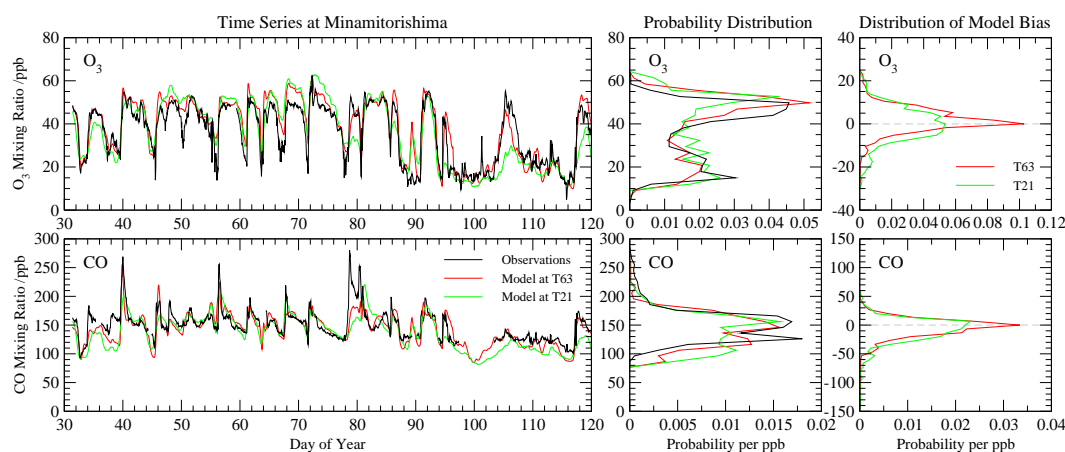


Fig. 2. Timeseries of hourly-mean surface O_3 and CO observations at Minamitorishima WMO/GAW station in February–April 2001 and FRSGC/UCI CTM results at T21 and T63 resolution. Right-hand panels show the probability distribution over the period and the distribution of the hourly model bias at each resolution.

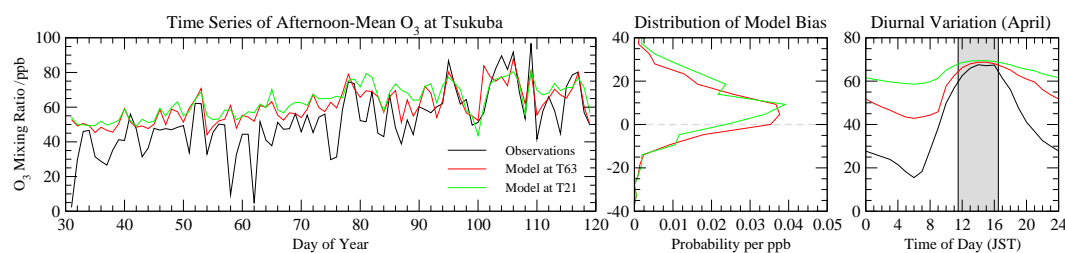


Fig. 3. Comparison of afternoon-mean O_3 observations (12:00–16:00 h JST) at Tsukuba WMO/GAW station (black) with CTM results at T21 (green) and T63 (red) resolution. Right-hand panels show the distribution of the hourly model bias and the monthly mean diurnal variation in observed and modelled O_3 in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

area. To minimize the influence of nighttime removal we compare afternoon concentrations (12:00–16:00 LT) 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 suburban sites where the efficiency of nighttime 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 and in fine-scale meteorological features such as the frontal banding seen in Fig. 2, 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

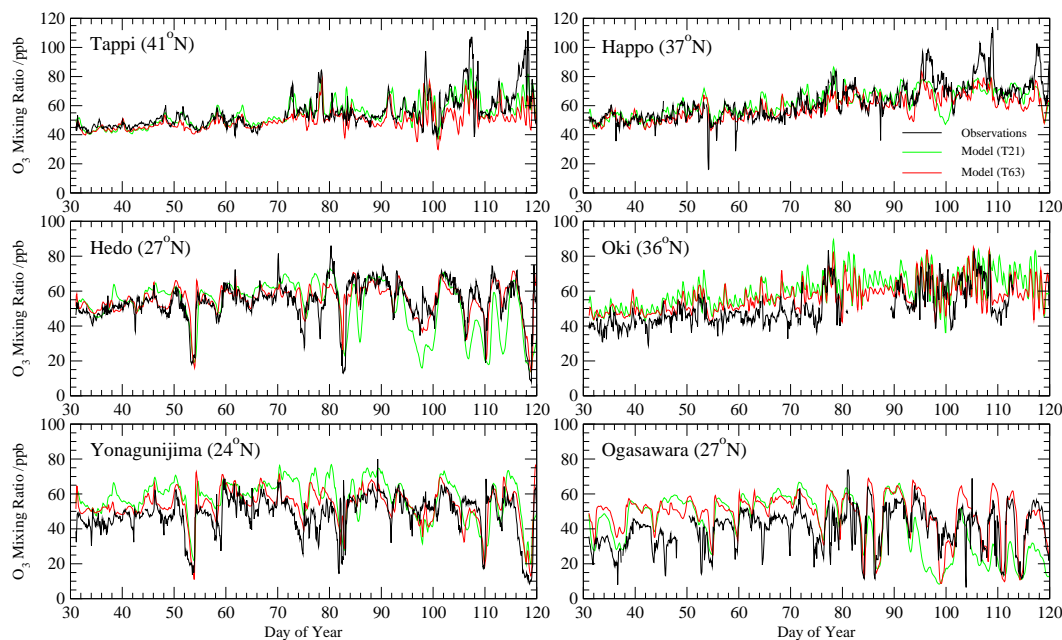


Fig. 4. Time series of hourly-mean O_3 observations at selected remote EANET and WMO/GAW measurement sites in February–April 2001 (black) and FRSGC/UCI CTM results at T21 (green) and T63 (red) resolution.

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

Site/Station	<i>n</i>	Mean O_3 ($\pm 1\sigma$)			Mean Bias		RMSE	
		Obs.	T21	T63	T21	T63	T21	T63
Remote								
Rishiri	1175	44.1 \pm 8.4	45.8 \pm 5.0	44.3 \pm 3.4	1.8	0.2	7.3	9.2
Tappi	2013	55.0 \pm 11.6	54.7 \pm 9.1	49.8 \pm 6.9	−0.3	−5.2	8.0	10.3
Happo	2087	62.8 \pm 12.9	62.1 \pm 8.9	58.7 \pm 8.2	−0.6	−4.1	9.8	9.1
Oki	1730	50.2 \pm 10.1	59.4 \pm 9.1	55.8 \pm 8.3	9.3	5.6	12.5	9.2
Ogasawara	2039	40.0 \pm 11.3	41.4 \pm 16.2	48.6 \pm 12.4	1.5	8.6	16.2	13.4
Hedo	2103	53.9 \pm 10.9	51.8 \pm 14.0	54.8 \pm 9.0	−2.1	0.9	11.9	7.1
Yonagunijima	2128	48.6 \pm 11.0	57.3 \pm 11.6	53.5 \pm 9.9	8.7	4.9	13.7	9.7
Minamitorishima	2096	35.7 \pm 13.0	35.5 \pm 14.7	37.7 \pm 14.0	−0.2	2.0	8.1	6.1
Rural								
Ryori	1903	44.9 \pm 9.4	55.4 \pm 8.9	54.4 \pm 7.5	10.5	9.5	13.2	12.0
Sadoseki	2103	48.8 \pm 15.9	59.5 \pm 9.0	52.8 \pm 8.1	10.8	4.0	17.8	13.6
Yusuhara	2126	35.1 \pm 10.6	60.7 \pm 9.3	57.6 \pm 8.4	25.6	22.6	27.8	24.7
Suburban ^a								
Tsukuba	445	51.8 \pm 18.0	63.2 \pm 9.2	60.6 \pm 10.2	11.4	8.8	18.0	15.8
Ijira	445	52.9 \pm 16.2	65.3 \pm 10.0	60.7 \pm 8.9	12.4	7.8	17.8	14.7
Banryu	445	53.5 \pm 12.9	66.7 \pm 11.1	61.0 \pm 8.9	13.2	7.4	16.1	12.0

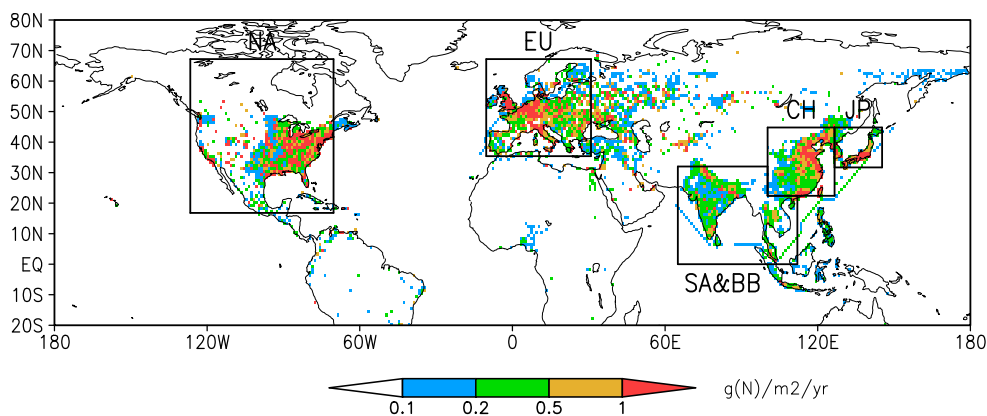
^a Only afternoon data (12:00–16:00 JST) used at suburban sites.

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; all

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

Site/Station	<i>n</i>	Mean CO ($\pm 1\sigma$)			Mean Bias		RMSE	
		Obs.	T21	T63	T21	T63	T21	T63
Ryori	1020	228.8 \pm 42.5	239.2 \pm 24.0	212.7 \pm 27.9	10.3	−16.1	41.7	42.5
Yonagunijima	1991	222.9 \pm 75.5	219.2 \pm 61.1	206.5 \pm 61.1	−3.7	−16.4	66.2	54.8
Minamitorishima	1896	149.7 \pm 26.3	140.1 \pm 27.3	144.6 \pm 25.2	−9.6	−5.1	23.2	18.0

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

locations lie in different grid boxes at T63. The following sections of this study focus on model simulations at T63 resolution only.

We conclude the model evaluation by examining the importance of the location of measurement sites. 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). 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 Source contributions over the Tokyo region

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 period was then repeated at higher resolution (T63), and the results from these runs are described here. This method provides a reasonable estimate

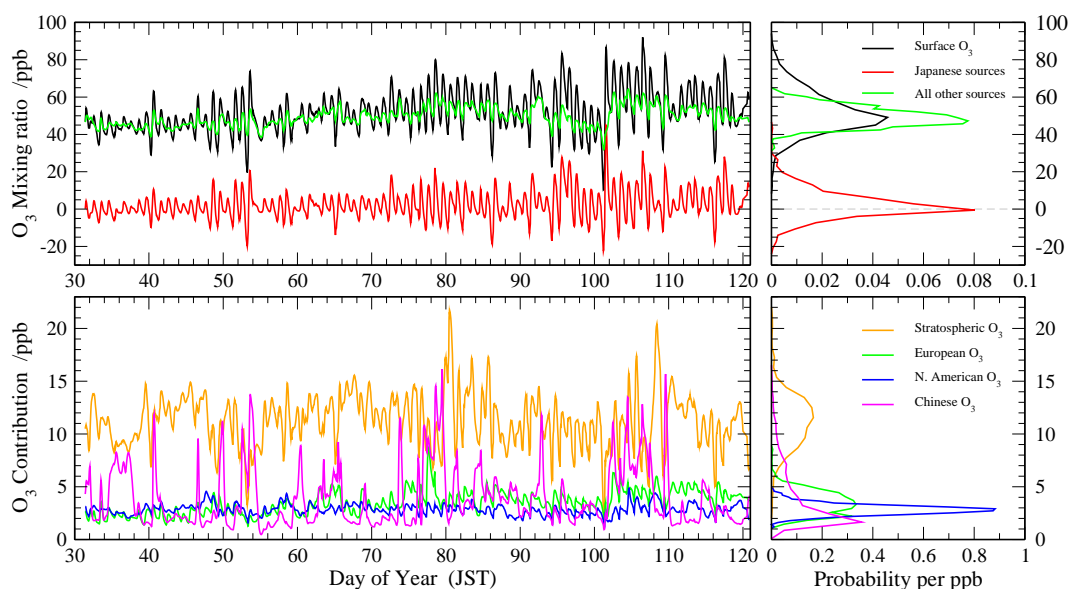


Fig. 6. Hourly mean surface O_3 mixing ratios in Tokyo in February–April 2001 from the FRSGC/UCI CTM showing (a) total surface O_3 (black), O_3 from Japanese sources (red) and the difference between them (representing O_3 from all other sources; green), and (b) contributions to O_3 from Chinese (magenta),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

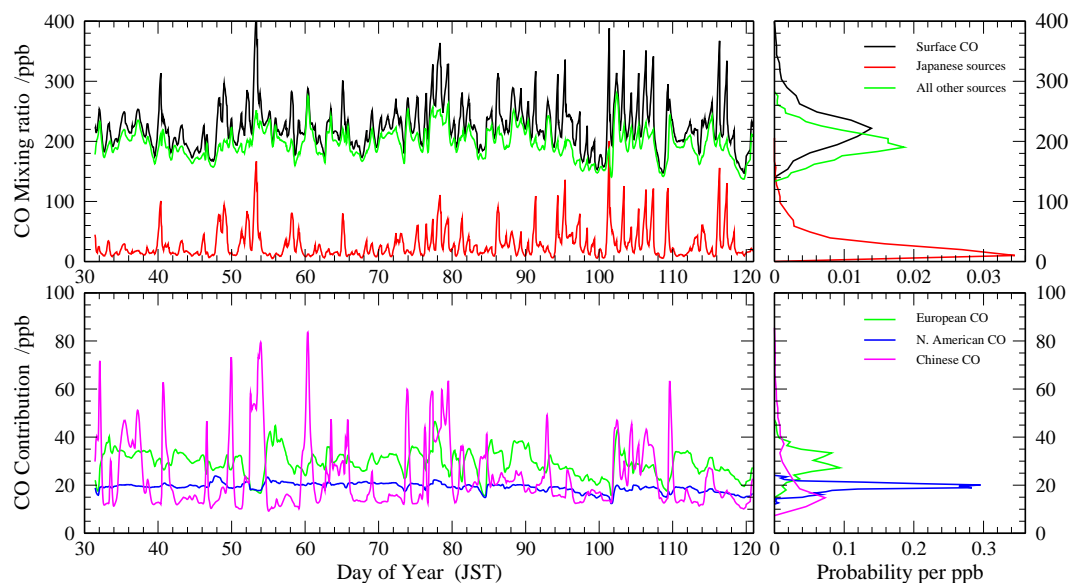


Fig. 7. Hourly mean surface CO mixing ratios in Tokyo in February–April 2001 from the FRSGC/UCI CTM showing (a) total surface CO (black), CO from Japanese sources (red) and background CO from other sources (green), and (b) contributions to CO from Chinese (magenta), European (green), and North American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

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

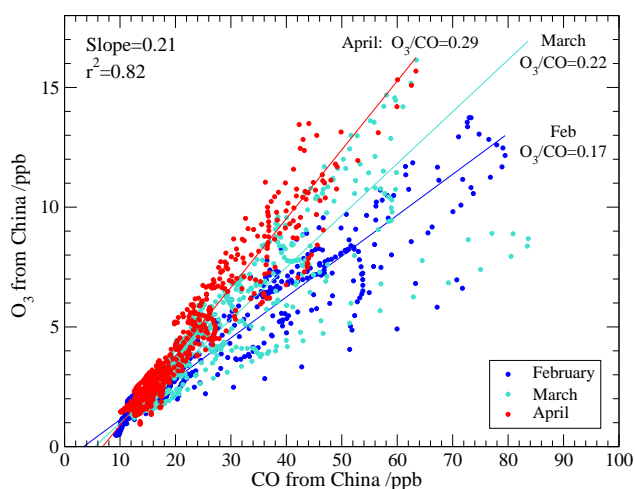


Fig. 8. Relationship between Chinese contributions to surface O_3 and CO over Tokyo revealing the increase in photochemical activity between February and April in 2001.

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 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).

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.

Hourly source contributions are shown in Fig. 9 as a function of the total surface O_3 at all times of day to demonstrate their influence during episodes of high O_3 over Tokyo. There

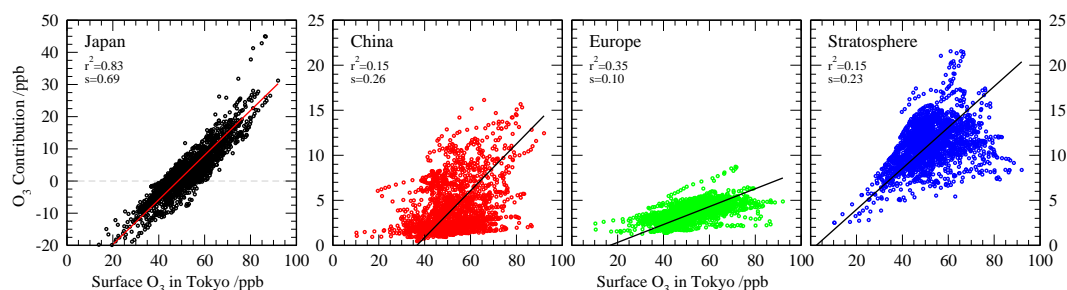


Fig. 9. Contributions of Japanese, Chinese and European industrial sources and transport from the stratosphere to hourly surface O₃ over Tokyo as a function of total modelled surface O₃. Graphs are labelled with the correlation coefficient (r) and slope (s).

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	

is a very high correlation between the Japanese contribution and total surface O₃ ($r^2 = 0.83$) indicating that local sources account for most of the variability in O₃ 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 O₃ exceeds the Japanese 1-h 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 O₃ 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 O₃ lies tightly between 15 and 33 % over the spring period, and although there is a notable decline when surface O₃ exceeds 50 ppb, the stratosphere still contributes an average of 12.2 ppb during O₃ episodes, very similar to the local Japanese contribution of 12.6 ppb.

During the highest episodes O₃ 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 O₃ peak, and industrial emissions from Chinese, European and North American sources together accounted for less than 10 %. How-

ever, during the 20 March (day 79) and 20 April (day 109) episodes Japanese sources account for only about 25 % of the O₃, 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. In contrast to 12 April, these two episodes were associated with large-scale anticyclonic influence over Northern Japan, and high O₃ was observed at Tappi and Happo at the same time (see Fig. 4). The remaining three episodes lie between these extremes, with Japanese sources accounting for about one third of O₃ 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 not greatly different during these periods, see Table 6. This suggests that trans-boundary and intercontinental transport contribute to O₃ levels over Japan during conditions that favour O₃ build-up, and may thus impact attainment of air quality standards in Japan.

Table 5. Source Contributions to Afternoon Surface O₃ in Tokyo During Spring 2001.

	Feb		Mar		Apr		Feb–Apr	
	ppb (%)		ppb (%)		ppb (%)		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	

Table 6. Source Contributions to Surface O₃ (ppb) in Tokyo During High O₃ Episodes (O₃>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

4 Transport pathways and meteorology

The large differences seen between high O₃ 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 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 O₃ 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 O₃ 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 O₃ differs from that of CO, as O₃ has a shorter lifetime and surface contributions are therefore more strongly influenced by subsidence from the free troposphere than horizontal transport. North American O₃ correlates relatively well with stratospheric O₃ suggesting that they arrive by similar pathways following transport and subsidence from the middle and upper troposphere. European O₃ correlates with

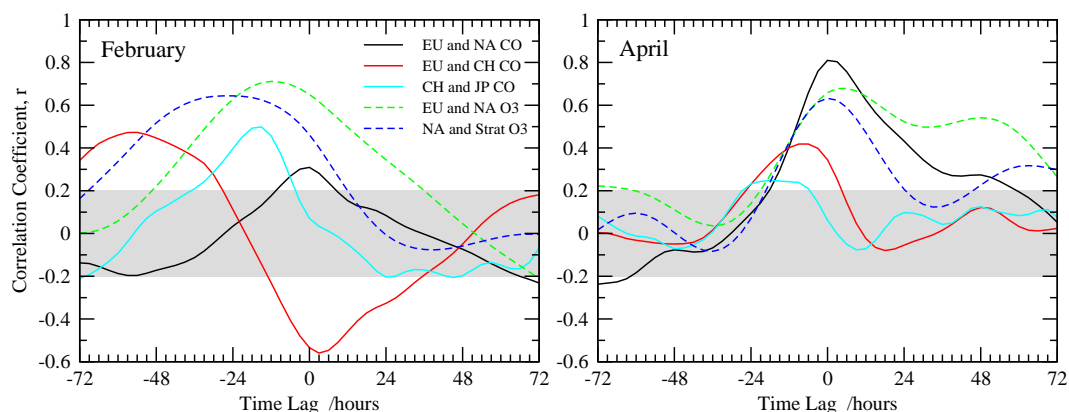


Fig. 10. Correlation between the contributions of key source regions to O₃ and CO over Tokyo in February (left) and April (right).

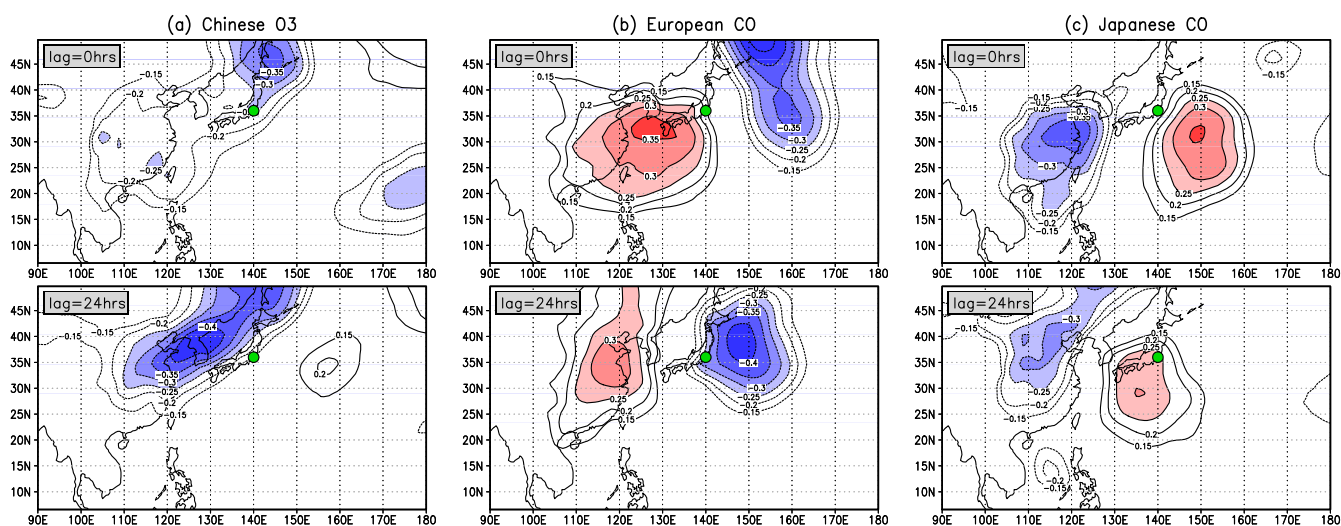


Fig. 11. Correlation between mean sea-level pressure and the contributions of (a) Chinese O₃, (b) European CO and (c) Japanese CO over Tokyo (marked with a circle) in spring 2001. Positive correlations are highlighted in red and negative correlations in blue. Lower panels show the correlation coefficient lagged by 24 h, and thus the relationship with mean sea-level pressure one day earlier.

stratospheric O₃ 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. We show examples for both O₃ and CO; the correlation patterns for both species are similar for Chinese sources, but the strong diurnal signal in O₃ 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

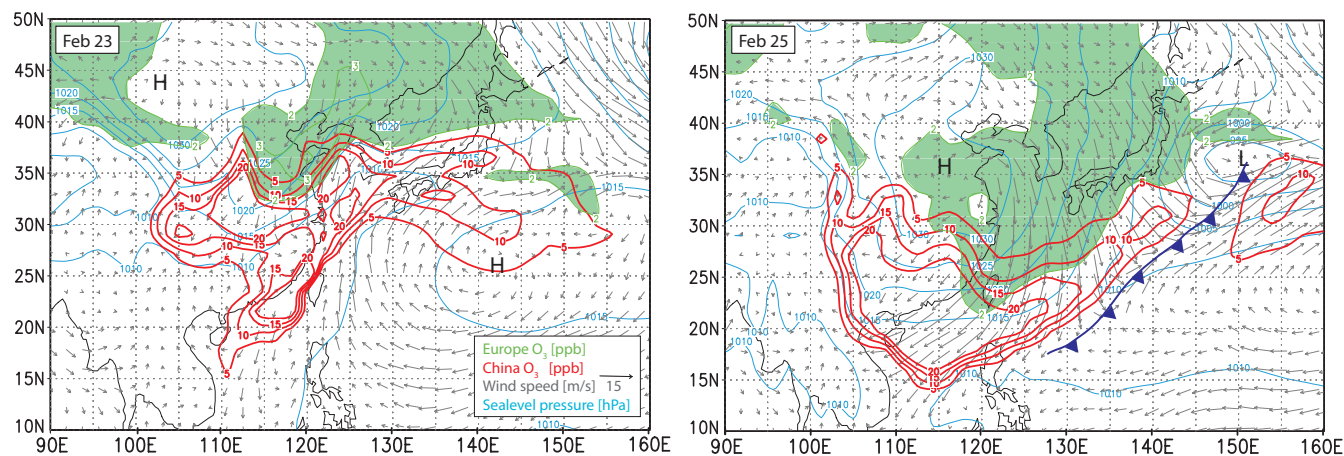


Fig. 12. Chinese and European contributions to O₃ and CO over Japan at 00:00 UTC on 23 and 25 February 2001 showing the development and movement of air masses associated with a developing frontal system (approximate position on 25 February marked in blue).

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 O₃ 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. 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 O₃ 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 O₃ (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 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 O₃ of 2.2 to 3.5 ppb. While there is some mixing of air influenced by

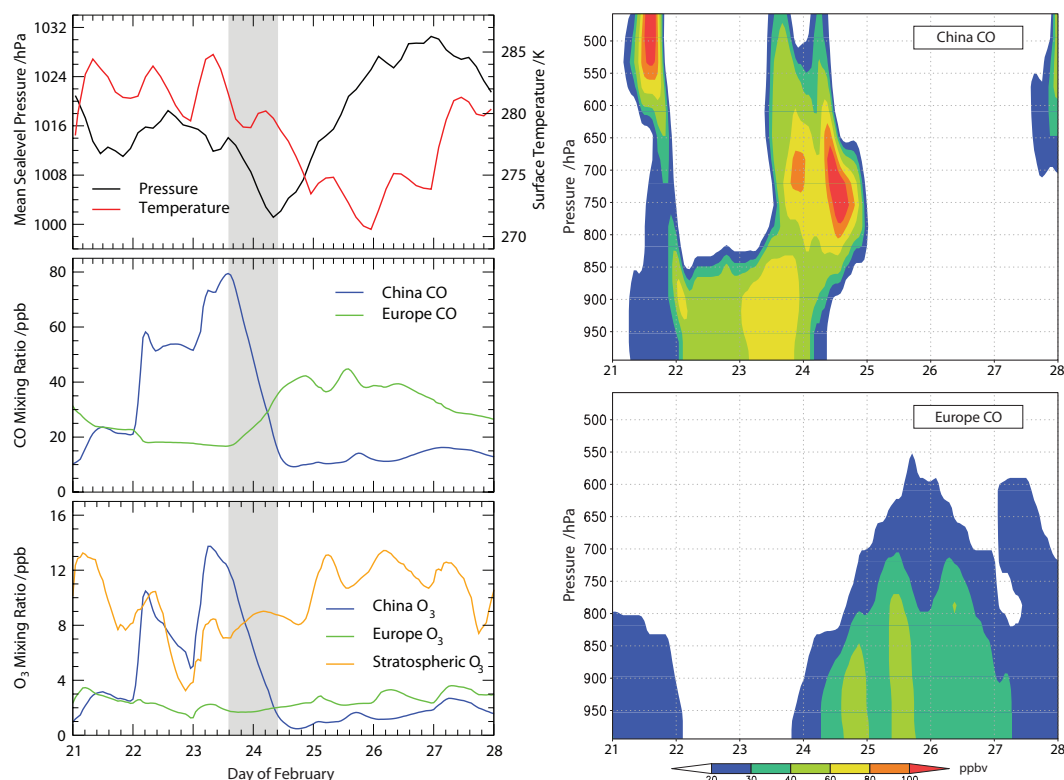


Fig. 13. Variations in surface pressure and temperature over Tokyo during the week of 21–28 February 2001, showing the passage of the frontal system shown in Fig. 12 (highlighted in grey) and the associated changes in the contribution of distant sources to O_3 and CO (left panels). Vertical profiles 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.

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 ppbv of O_3 and 100 ppbv 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 ppbv and CO reaching 180 ppbv, 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 ppbv of stratospheric O_3 which persist for about a day. At the surface, stratospheric contributions average 12 ppbv in the air behind the front compared with about 6 ppbv 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 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 ppbv for an 8-h period (for 5 h in the observations) and peaks at more than 70 ppbv. Although early in the year, the fair-weather anticyclonic conditions contribute to substantial local O_3 production (20 ppbv from Japanese sources) and the southwesterly flow brings as much as 14 ppbv 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 O₃ 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 O₃ 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 O₃ in this season due to strong O₃ removal at nighttime over polluted emission regions. Distant anthropogenic precursor emissions over China, Europe and North America make small but significant contributions to surface O₃ (7 %, 6 %, and 5 % respectively), but together these sources make up 19 % of O₃ over Japan, exceeding 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 O₃ 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 O₃ 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 O₃ over Tokyo in springtime. While the contribution from European and North American sources 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 O₃ over Tokyo in spring, contributing to the observed trend. The magnitude of this O₃ 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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