On the regional background levels of carbon monoxide observed in East Asia during 1991~2004

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Abstract The carbon monoxide (CO) concentrations observed in Mt. Waliguan in China (WLG), Ulaan Uul in Mongolia (UUM), Tae-ahn Peninsula in Korea (TAP) and Ryori in Japan (RYO) were analysed between 1991 and 2004. The average annual concentration of CO, a toxic air pollutant, was the highest at TAP (233 ± 41 ppb), followed by RYO (171 ± 36 ppb), UUM (155 ± 26 ppb) and WLG (135 ± 22 ppb). CO tends to be highest in spring and lowest in summer in East Asia, with the exception of WLG. TAP had the highest CO concentrations in all seasons compared with WLG, UUM and RYO and displays a wide short-term variability in concentration. This is caused by large-scale air pollution owing to its downwind location, close to continental East Asia. CO concentrations observed at TAP were analysed as follows: according to the origin of the isentropic backward trajectory and its transport passage, as continental background airflows (CBG) measuring 216 ± 47 ppb, regionally polluted continental airflows (RPC) measuring 316 ± 56 ppb, oceanic background airflows (OBG) measuring 108 ± 41 ppb and partly perturbed oceanic airflows measuring 161 ± 6 ppb. The high concentrations of CO at TAP are because of the airflow originating from the East Asian continent, rather than the North Pacific. RPCs, which pass through eastern China, appear to have high CO concentrations in spring, autumn and winter. OBGs, however, are affected by North Pacific air masses

with low CO concentrations in summer. National Oceanic and Atmospheric Administration satellite images and the Goddard Earth Observing System (GEOS)-Chem simulation model confirmed that a large-scale air pollution event originated in eastern China and reached the Korean Peninsula and the Korea East Sea by way of the Yellow Sea. It is noteworthy that the overall trend at TAP does not show an increase despite the fact that energy use in China approximately doubled from 1991 to 2004.

Keywords Carbon monoxide · Regional background level · Large-scale air pollution · Transport passage · Satellite image

Introduction

The East Asian region, which includes China, Korea and Japan, is one of the world's three top manufacturing regions and has experienced consistent annual increases in anthropogenic emissions (Streets et al. 2001). Total emissions of CO for the Asian region in 2000 were estimated as 279 Tg, of which 116 Tg was from China, accounting for 42% of emissions (Streets et al. 2003). Although 90–95% of CO emissions are removed through their reaction with OH in the atmosphere (Logan et al. 1981), toxic CO is widely distributed in the East Asian region by long-range transport, owing to the large quantity of emissions and their several-months lifetime, depending on the season (Novelli et al. 1998).

Air pollutants emitted on a large-scale from the East Asian continent, particularly from China, are recognised to affect Korea, some originating from a distance of several hundred or even thousands of kilometres (Chung et al. 2001, 2003; Kim and Chung 2005). Emissions in East Asia also affect ground measurements of CO in regions such as

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Japan, the North Pacific and the west coast of North America (Akimoto et al. 1996; Jaffe et al. 1997; Liang et al. 2004; Pochanart et al. 2004). Intensive observation campaigns have therefore been conducted to identify the atmospheric chemical components in the East Asian region and the characteristics of their movements (Hoell et al. 1996, 1997; Huebert et al. 2003; Jacob et al. 2003).

Long-term observation of background sites yields valuable information regarding changes in atmospheric components caused by long-range transport. Background concentrations of CO have remained approximately constant in time, with systematically higher concentrations in the Northern Hemisphere compared with the Southern Hemisphere, displaying the highest concentrations in the high latitudes of the Northern Hemisphere between late winter and early spring (Novelli et al. 1998). This study, however, focuses on the regional background concentration of CO based on long-term observations in East Asia, which is mainly affected by westerly winds high in CO emissions. In general, CO measurements at TAP were made when the fresh air arrived, e.g., with the arrival of a new anticyclone. Thus, high CO values greater than 300 ppb were not included in this study. Regional background concentrations of CO at Tae-ahn on the Korean Peninsula, adjacent to the East Asian continent, were measured in the early 1990s (Chung and Lee 1993). The characteristics of regional background concentrations of CO collected from 1991 to 2004, according to the transport passage of the airflows in the East Asian region, are analysed in this study.

Data and analysis methods

Figure 1 shows the geographic locations of the measurement sites in the East Asian region. The National Oceanic

Fig. 1 Locations of the background sites in the East Asian region. Passages of the movement of the airflows arriving at TAP are classified as CBG transport, RPC transport, OBG transport and PPO transport

Table 1 Details of the background sites in the East Asian region

Station Name	Country	Latitude	Longitude	Altitude (a. s. l)
WLJ	China	36°17' N	100°54' E	3,810 m
UUM	Mongolia	44°27' N	111°05' E	914 m
TAP	Korea	36°43' N	126°07' E	20 m
RYO	Japan	39°02' N	141°49' E	260 m

and Atmospheric Administration Earth System Research Laboratory (NOAA/ESRL) Carbon Cycle Greenhouse Gases Group in the USA has a globally distributed air-sampling network in the East Asian region, which includes TAP in Korea, UUM in Mongolia and WLJ in China. The Japan Meteorological Agency operates the RYO observatory. Further details pertaining to each observation site are summarised in Table 1. TAP and RYO, as opposed to WLJ and UUM, are located on the downwind side of the East Asian continent, and TAP is most affected by anthropogenic air-pollutant emissions, which arrive by long-range transport from China (Chung et al. 2001; Kim and Chung 2005). Air for measuring CO concentrations was collected by equipment provided by the NOAA/ESRL, which pumped air into two (duplicate) glass flasks. Measurement was conducted once or twice each week at 12–16 Local Standard Time (LST), beginning in 1990. The collected air samples were sent to the NOAA/ESRL in the USA to obtain accurate concentrations of CO. CO₂ and CH₄ concentrations were also analysed, and the regional background concentrations of these gas components observed at TAP were studied (Chung and Lee 1992; Lee and Chung 1993). A detailed analysis of the collected air is described in Novelli et al. (1998), using the CO concentration data from TAP, WLJ and UUM, analysed by the NOAA/ESRL for 1991 to 2004. CO concentrations at RYO are based on data from the Japan Meteorology Agency. In this study, CO data measured at Mauna Loa (MLO) in Hawaii by NOAA/ESRL are also compared with the data from four Asian stations.

To estimate the regional background CO concentration along the airflow transport passages in the East Asian region, CO concentrations and isentropic backward trajectories measured at TAP between 1991 and 2004 were analysed. The Hybrid Single-Particle Lagrangian Integrated Trajectory model (Draxler and Rolph 2003) was used to track the path of airflows in the East Asia region. The trajectories arrived at 09 LST on each day when the air was collected at TAP.

The isentropic backward trajectories arriving at TAP, originating approximately 1,000 km away, are divided into four groups according to their origin and transport routes

(Fig. 1). The trajectory passages that circulate during transport are excluded from the analysis. Observed CO data in the East Asian region demonstrate clear seasonal changes owing to the influences of Siberian continental air masses affected by the monsoon and oceanic air masses from the North Pacific. Isentropic backward trajectories are accordingly divided into continental- and oceanic-originating airflows.

The trajectories of air masses of continental origin are divided into two groups depending on the distribution of CO emission in the East Asian region (Liang et al. 2004) and the transport passage before it reaches TAP. The isentropic backward trajectory of continental origin is classified as follows: continental background (CBG) transports originating in northeast China and the eastern Siberian region. Both pass through the NW~NNE portion of the Korean peninsula, and regionally polluted continental (RPC) transports, originating from the Chinese continent and landmasses at lower latitudes, pass through the eastern area, then move south to Bohai Bay in China and the Yellow Sea before arriving at TAP. CBGs do not pass through the area where there are high industrial CO emission rates. RPCs pass through the industrial district in the eastern region of China. This region also includes the densely populated urban areas of Beijing, Tianjin and Shanghai.

Kim and Chung (2005) classify the ozone concentrations measured at TAP into CBG concentrations and the level of regionally polluted airflows, according to the isentropic backward trajectories. Continental airflows appear mainly in spring, autumn and winter. Trajectories of oceanic origin are classified by oceanic background (OBG) transports originating in the western North Pacific, arriving at TAP by way of the East China Sea and the Yellow Sea, and partly perturbed oceanic (PPO) transports passing through Japan and Korea.

Results and discussion

CO distribution in East Asia

Figure 2 shows the annual average variations in CO concentration measured at WLG in China, UUM in Mongolia, TAP in Korea, RYO in Japan and MLO in Hawaii between 1990 and 2004. Compared with CO data of MLO in Hawaii (19.54° N, 155.58° W, 3,397 m), with 96 ± 19 ppb of CO, the East Asian region shows high concentrations of 135 ± 22 ppb at WLG, 155 ± 26 ppb at UUM, 171 ± 36 ppb at RYO, and 233 ± 41 ppb at TAP. Although TAP and WLG are in close latitudinal proximity, WLG is at high altitude and upwind of the East Asian industrial region. TAP appears to display the highest carbon monoxide concentrations, being located 350–700 km downwind of the eastern part of China. UUM, located upwind of where the continental pollution originates, demonstrates lower carbon monoxide concentrations than at TAP but higher concentrations than at WLG. It should be noted that annual mean CO at TAP appears to be trending down slightly over these last 14 years, despite the rapid industrialisation of China.

RYO in Japan, in an easterly direction from the East Asian continent further downwind than TAP, displays lower concentrations than those at TAP. In terms of annual mean variations, UUM, TAP, RYO and MLO were observed in 1998 to have high CO concentrations. The annual mean of ozone concentrations was also high at Chongwon, an agricultural area in central South Korea (Kim and Chung 2003). More forest fires in Siberia occurred in 1998 than in any other year on record, and Kato et al. (2002) suggest that CO concentrations and the ozone concentrations measured at Happon, Japan in 1998 had increased by ~ 30 and ~ 7 ppb, respectively, as a result of the long-range transport caused by the forest fires in Siberia. At that time, the CBG trajectories stayed at low altitudes moving to the east.

Fig. 2 Annual mean CO concentrations at Mt. Waliguan (WLG) in China, Ulaan Uul in Mongolia (UUM), Tae-ahn Peninsula (TAP) in Korea, Ryori (RYO) in Japan, and Mauna Loa (MLO) in Hawaii for 1990–2004

Figure 3 shows histograms of the CO concentrations measured at TAP, UUM and WLG between 1991 and 2004. These three measurement sites display important differences. The limited range of the histogram in the

Fig. 3 Histograms of CO concentrations measured at TAP, UUM, and WLG for 1990–2004

graph for WLG explains the consistently similar CO concentrations. CO data at TAP demonstrate, however, a wide range of distribution because CO is affected by large-scale transport of air pollution (LSTAP). The distribution of CO concentrations at UUM falls between those for WLG and TAP. The bump at low CO concentrations is notable for TAP but not for the other sites, which confirms the occurrences of transport from the OBG sector.

Seasonal variation of CO concentrations in East Asia

Figure 4 shows the monthly average variations in CO concentration observed at WLG, UUM, TAP, RYO and MLO between 1990 and 2004. Generally, CO concentrations are highest between March and May and lowest in July and August (November at WLG), and gradually increase in autumn and winter. The CO characteristics of the East Asian region reflect the emissions and their long-range transport, influenced by continental and oceanic air masses, with similar results to observations made at various sites in Japan (Pochanart et al. 1999, 2004). CO from fossil fuel combustion accumulates in the atmosphere in winter and early spring, increasing CO concentrations. In summer, however, the region is influenced by maritime airflows originating in the Pacific Ocean. These maritime air masses generally have low CO concentrations.

The differences between the highest and the lowest monthly mean CO concentrations were 85 ppb at TAP and 92 ppb at RYO, suggesting significant seasonal variations. The variation is larger at RYO than at TAP because the area is under the influence of North Pacific air masses during summer. WLG, however, displays a minimum of about 110 ppb in November, after a maximum of about 158 ppb in March. Seasonal variations at WLG compare to measurements at TAP. Both observation stations are located at similar latitudes. WLG measurements displayed a small variation compared with the other regions because it is located upwind of East Asia and is less influenced by the atmospheric boundary layer, located at an altitude of 3,810 m. The difference between the highest and the lowest monthly average concentrations of CO at UUM was 54 ppb, which falls between the measurements obtained at WLG and TAP.

Figure 5 depicts the seasonal histogram of CO concentrations measured at TAP, UUM and WLG between 1991 and 2004. Spring in this region covers March to May, summer June to August, autumn September to November and winter December to February. A significant difference of maximum distributions can be observed between spring and winter in the histogram of these three areas. The sharp and narrow distribution graphs at WLG and UUM in all seasons show almost constant CO concentrations and are

Fig. 4 Monthly mean CO concentrations at WLG, UUM, TAP, RYO, and MLO for 1990–2004

similar in summer and autumn. The wide distribution of CO concentrations at TAP, however, reflects the large-scale air pollution. Many cases of high and low concentration were also observed at TAP in summer compared with other seasons.

CO concentration levels according to trajectory categorisation

The cases with clear transport passages according to their origin and transport routes, originating approximately

Fig. 5 Histograms of CO concentrations measured at TAP, UUM, and WLG for 1990–2004: Spring, summer, autumn and winter are March–May, June–August, September–November, and December–February, respectively

1,000 km from TAP, were 242 of 526 days, classified by four categories. Representative cases of each category are shown in Fig. 6. The backward trajectory that originated in Siberia arrived from the NW~NNE at the Korean Peninsula, moving past the eastern part of Inner Mongolia and Manchuria at a high altitude of more than 1.5 km. Such CBG is high in frequency from the NNW~NW at 68%. RPC reaches TAP from the Siberian continent by way of Mongolia, the inland and eastern parts of China and the Yellow Sea, showing 19%. The trajectories originating in the North Pacific move at low altitudes below 1 km; 11% of the OBG reaching TAP by way of the East China Sea and the Yellow Sea does not pass through the emission-producing areas and 2% of PPO passes through Korea and Japan.

Table 2 shows the CO concentration levels at TAP according to categories of isentropic backward trajectories. The concentrations of CO by trajectory that originates on the East Asian continent reaching TAP are higher for the RPC sector (316 ± 56 ppb) than for CBG (216 ± 47 ppb), and the seasonal variation is higher for RPC than for CBG through all seasons. Concentrations of CO for RPC are highest in spring, which is similar to the seasonal variation in Fig. 4. The passage of trajectories that originate in the North Pacific and arrive at TAP is measured mainly in summer and partially in autumn. The weighted average of the non-RPC sectors is 200 ppb, so that the RPC sector average is 115 ppb higher. If the CO emissions in China had increased in proportion to its energy consumption, we would have expected the average CO concentration in the

Table 2 Average CO concentrations of CBG, RPC, OBG and PPO, (unit: ppb)

Month	CBG	RPC	OBG	PPO
Mar~May	236 ± 41	327 ± 44	–	–
Jun~Aug	–	–	101 ± 35	161 ± 6
Sep~Nov	193 ± 50	300 ± 80	187 ± 3	–
Dec~Feb	223 ± 42	315 ± 42	–	–
Average	216 ± 47	316 ± 56	108 ± 41	161 ± 6

RPC sector to increase from about 275 ppb (75 above 200) to 350 ppb (150 above 200). We see no evidence of that, which suggests that CO emissions as a fraction of total combustion have decreased significantly.

OBGs passing over the ocean through the East China Sea and the Yellow Sea have lower concentrations of CO (108 ± 41 ppb) than PPO (161 ± 6 ppb), which passes through Japan or Korea. The concentrations of CO in oceanic airflows are lower than those of continental origin because the North Pacific air masses contain low levels of CO. CO reacts with OH, which removes it from the atmosphere, and concentrations of CO in the atmosphere can be partly affected by the seasonal variation of OH (Novelli et al. 1998). The CO concentrations in East Asia, however, are more strongly affected by the spatial distribution of emissions, long-range transport and variations in emission rates.

Large-scale transport of air pollution in the Yellow Sea region

Fig. 6 Four examples of airflows arriving at TAP show the isentropic backward trajectories: CBG transport, RPC transport, OBG transport and PPO transport

Figure 7 analyses NOAA satellite images and meteorological maps and employs the Goddard Earth Observing System (GEOS)-Chem model (Bey et al. 2001) to examine individual cases of LSTAP (Chung et al. 2001; Chung et al. 2003). The NOAA satellite image in Fig. 7a shows the LSTAP originating in the area near Shanghai down the Yangtze River in the southeastern part of China and expanding to the Korea East Sea by way of the Yellow Sea and the middle part of the Korean Peninsula. In the surface weather map (Fig. 7b) at 21 LST on December 9th, which is about 15 h earlier than the NOAA satellite image, the middle part of the Korean Peninsula, facing the cold front of the migrating cyclone from the north–west, started to be affected by the south–west airflow from southeastern China. The synoptic feature lasted until December 10th. Such a synoptic feature pattern, which transports large-scale air pollution from the Yellow Sea region, specifically from the eastern part of China to the Korean Peninsula, was also observed in a previous study (Kim and Chung 2005).

The transport of large-scale air pollution observed in the NOAA satellite image coincides with the distribution of CO concentration simulated at 930 hPa (700 m) by the GEOS-

Fig. 7 **a** Satellite image of NOAA 17 (1141 LST, 10 Dec 2004) showing a large-scale transport of air pollution (*LSTAP*) over the Yellow Sea, Korea and the Korea East Sea. **b** A meteorological map showing the movement of airflow from south-east China to the Korea East Sea. **c** Model simulation of CO concentrations at 930 hPa

Chem model (Fig. 7c). At 930 hPa, the region near Shanghai shows a concentration range of over 350 ppb and the concentration range of the region that expands to the Korea East Sea toward the north-east through the Yellow Sea and the Korean Peninsula is over 300 ppb, which is observed in the shape of a large-scale pollution mass, as shown in the NOAA satellite image. Figure 7d is the meridional average of the concentration of CO according to altitude, in the region of 28–48° N latitude and 113–135° E longitude, based on the GEOS-Chem model. A high concentration range of an average of over 250 ppb was observed above the eastern part of China and the Korean Peninsula. While the intervals between the isograms become larger in altitudes over 3 km, the isograms of 125–200 ppb CO concentration are concentrated at an altitude of 1–3 km, which indicates the altitude distribution in LSTAP.

(700 m) and **d** Meridional average (latitude, 28–48°N; longitude, 113–135°E) of CO concentrations conducted by use of GEOS-CHEM global three-dimensional model of tropospheric chemistry (<http://www-as.harvard.edu/chemistry/trop/geos/>)

Conclusions

An analysis of a long-term observation of CO concentration conducted at WLG in China, UUM in Mongolia, TAP in Korea and RYO in Japan between 1991 and 2004 was made to analyse the regional background concentration level of CO in East Asia. CO values in these areas are high in annual average concentrations of CO, in the order of WLG 135 ± 22 ppb, UUM 155 ± 26 ppb, RYO 171 ± 36 ppb and TAP 233 ± 41 ppb. In terms of seasonal variation of CO concentration, all the areas except WLG are high in spring and low in summer, which is similar to observations made in other areas in Japan. WLG and UUM are geographically at a similar latitude to TAP, and RYO is downwind of the East Asian continent. TAP, located 350–700 km downwind of China, is under greater influence of large-scale air pollution than the other observation sites in this study.

The CO concentration at TAP on the Korean Peninsula can be divided into four categories according to an analysis of the isentropic backward trajectories. The concentrations of CO of CBG and RPC of East Asian origin were 216 ± 47 and 316 ± 56 ppb, respectively. OBG, which originates in the low-latitude North Pacific and travels above the ocean, was 108 ± 41 ppb, and PPO, which passes through Korea or Japan, was 161 ± 6 ppb. The concentration of CO in an airflow originating in high-latitude Siberia and affecting TAP is higher than that of the passage from the low-latitude North Pacific. In the East Asian region, the case analysis using the NOAA satellite image proves that TAP is being affected by the large-scale transport of air pollution from China.

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