

**Evaluation of
anthropogenic
emissions of carbon
monoxide**

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**Evaluation of anthropogenic emissions of
carbon monoxide in East Asia derived
from observations of atmospheric
radon-222 over the Western North Pacific**

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We used the observed $\text{CO}/^{222}\text{Rn}$ ratio in Asian outflows at Minamitorishima (MNM), Yonagunijima (YON), and Ryori (RYO) over the Western North Pacific from 2007 to 2011, together with a three-dimensional chemical transport model (STAG), in order to estimate anthropogenic emissions of CO in East Asia. The measurements captured high-frequency synoptic variations of enhanced ^{222}Rn (ERN) events associated with long-range transport of continental air masses. ^{222}Rn and CO showed high correlation during the ERN events observed at MNM and YON in the winter and spring, but not at RYO. The STAG transport model reproduced well the concentration of observed ^{222}Rn when forced with constant and uniform flux density of $1.0 \text{ atom cm}^{-2} \text{ s}^{-1}$, but underestimated the associated enhancement of synoptically variable CO caused by the underestimated flux values in the EDGAR ver. 4.1 emission database used in the model for East Asia. Better estimates for the East Asian emission were derived using a radon tracer method based on the difference in the enhancement ratio of $\text{CO}/^{222}\text{Rn}$ between observation and model. The anthropogenic emission of CO for China, Japan, and Korea was estimated to be $203 \text{ Tg CO yr}^{-1}$, 93% of which originated in China. When compared with other estimated emissions of CO, our estimated result showed consistency with those of the inverse method, whereas the emission database of EDGAR was about 45% smaller than our anthropogenic estimation for China.

1 Introduction

Recently, East Asia has become one of the large source regions of anthropogenic pollutants, along with North America and Europe. The emissions from East Asia continue to increase due to the growing economy and population (Akimoto, 2003). The continental outflow of pollutants from East Asia has had a significant impact on the environment in the downwind regions of the Western North Pacific (Tanimoto et al., 2005). Several studies have also provided evidence for various influences on the air quality over

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certain regions of North America due to the long-range transport of polluted air masses from Asia (e.g., Andreae et al., 1988; Parrish et al., 1992; Husar et al., 2001).

The anthropogenic emission of trace gases from East Asia is not well quantified. In particular, emission estimates of carbon monoxide from incomplete combustion are characterized by large uncertainties due to variability in the burning of fossil fuel, bio-fuel, and biomass in space and time. Bottom-up estimates based on the socioeconomic data have produced several emission databases, such as those of Streets et al. (2003), the Intercontinental Chemical Transport Experiment-Phase B (INTEX-B) emission inventory (Zhang et al., 2009), the Regional Emission inventory in Asia (REAS) (Ohara et al., 2007), and the Emission Database for Global Atmospheric Research (EDGAR) (Olivier et al., 2005). Another method to estimate surface emissions is to use a top-down approach, such as an inverse method (Heald et al., 2004; Kopacz et al., 2009; Hooghiemstra et al., 2011). However, there are significant disagreements between the bottom-up and top-down emission estimates for East Asia (Heald et al., 2004; Palmer et al., 2006). Those disagreements are caused by substantial uncertainties associated with insufficient knowledge about the source regions and emission factors used to convert the socioeconomic data into emission values in the bottom-up approach, and by uncertainties in the atmospheric transport mechanism and the lack of sufficient observational data to constrain the model in the inverse method. In this study we propose a third, and independent, method to assess and evaluate the disagreement.

Radon tracer method has been used to estimate continental emissions of methane (CH_4) (Thom et al., 1993; Schmidt et al., 1996; Levin et al., 1999), nitrous oxide (N_2O) (Schmidt et al., 2001), carbon dioxide (CO_2) (Schmidt et al., 2003; Hirsch, 2007), Chlorofluorocarbons (CFCs) (Biraud et al., 2000, 2002) in Europe. Radon-222 (^{222}Rn) has been used to parameterize transport and dilution of ground level emissions of trace gases in the atmospheric surface layer (Kuhlmann et al., 1998; Schmidt et al., 2001, 2003). ^{222}Rn is a radioactive noble gas with a half life of 3.824 days (e-folding time $\tau = 5.52$ days) and is produced by the decay of radium-226 distributed uniformly in the soil. ^{222}Rn exhalation from soils has been found to be rather homogeneous in

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a restricted region and almost constant with time (Dörr and Münnich, 1990). Exhalation rate from ocean is about two orders of magnitude less than that from soils (Wilkening, 1975). Thus, ^{222}Rn is a useful tracer for air masses originating from land. Our study is a first one to utilize observed ^{222}Rn in the radon tracer method to obtain emission estimates of trace gases for East Asia.

The National Institute of Advanced Industrial Science and Technology (AIST) and the Meteorological Research Institute (MRI) have developed a high precision ^{222}Rn measuring system (Wada et al., 2010) and used it to measure atmospheric concentration of ^{222}Rn on the ground for more than three years at remote sites in the Western North Pacific. In addition, the Japan Meteorological Agency (JMA) has been simultaneously measuring atmospheric concentrations of trace gases such as carbon monoxide (CO), carbon dioxide (CO_2), methane (CH_4), and ozone (O_3) at the same sites for more than a decade (Tsutsumi et al., 2006; Wada et al., 2007, 2011). Wada et al. (2011) showed that the combustion-related enhancement ratio of CO to CO_2 observed at these sites depended on the emission ratio of CO to CO_2 in East Asia. In addition, Wada et al. (2010) demonstrated that the enhanced ^{222}Rn peaks observed at MNM showed high correlation with synoptic scale variation of CO observed simultaneously at this station. These results indicated that the emission ratios of the trace gases emitted from East Asia are conserved in the enhancement ratios observed at these sites. Thus, emissions of the trace gases in East Asia can be estimated from the observed data over the Western North Pacific by the enhancement ratio of CO and ^{222}Rn .

In this study, we show observational results of the atmospheric ^{222}Rn and CO at the remote sites in the Western North Pacific and resolve the relationship between ^{222}Rn and CO. Next, an estimation technique based on the radon tracer method is developed to obtain East Asian CO emission estimates based on the observed atmospheric ^{222}Rn . Finally, we compare our estimated results with other estimated values by the bottom-up and top-down methods.

2 Sampling and analytical methods

2.1 Sampling sites

JMA is operating atmospheric monitoring stations at Minamitorishima (24.3° N, 154.0° E, 8 m a.s.l.), Yonagunijima (24.5° N, 123.0° E, 30 m a.s.l.), and Ryori (39.0° N, 141.8° E, 260 m a.s.l.) under the Global Atmosphere Watch programme of the World Meteorological Organization (WMO/GAW) (Fig. 1).

Minamitorishima (MNM) is a remote coral island in the Western North Pacific, about 2000 km southeast of Tokyo. At this site, easterly winds associated with maritime air masses prevail much of the year, except in the winter season, when continental air masses from East Asia are transported by synoptic scale weather systems. Since this island is very small, about 1.4 km² with a population of about fifty people, the influence of local sources and sinks on the trace gas measurement is negligible (Wada et al., 2007).

Yonagunijima (YON) is a remote island located about 111 km east from Taiwan. At this site, northeasterly winds associated with the continental high pressure system prevail, except in the summer when southerly wind associated with the maritime air masses dominates the site. Although the island is located at the same latitude as MNM, YON is situated much closer to downwind of the Asian continent (Fig. 1), thus frequently influenced by the polluted air masses from Asia (Tsutsumi et al., 2006). Since the island is small, less-populated and lightly-trafficked, the local influence on the measurements is not significant.

Ryori (RYO) is located on the east coast of northeastern part of Japan. Northwest-erly winds from the Siberian high pressure system dominate the site throughout the year. In the summer season, however, southerly winds associated with the Pacific subtropical high pressure system are often observed. The station is located on the ridge of a hill along the sawtooth coastline facing the Pacific Ocean, at an elevation of 260 m a.s.l. Due to its geographical location and complex terrain around the station,

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RYO is influenced by local sinks and sources, along with long-range transport of maritime and continental air masses.

2.2 Measuring method

The ^{222}Rn analyzer used in this study was developed by AIST and MRI (Wada et al., 2010) for precise measurements of the atmospheric ^{222}Rn concentration. It is based on the electrostatic collection method which collects positively charged progenies of ^{222}Rn in the sampling air on the PIN photodiode for an alpha particle detector charged high negative voltage. It automatically allows continuous measurement, to obtain hourly mean data. Calibration of this analyzer was made at Nagoya University which detail method is described elsewhere (Iida et al., 1991, 1996). The detection limit of this measuring system is 0.16 Bq m^{-3} for hourly mean data.

The measurement by this ^{222}Rn analyzer has been made at MNM since September 2007, and at YON since November 2008. Our current sampling method is a commonly used one for the greenhouse gas observation, and is described in Wada et al. (2007, 2010). Air is sampled at the top of a 10 m observational tower. After drying by the electric cooling units and membrane tube dryers, the sample air is introduced into the ^{222}Rn analyzer at the flow rate of 3.0 l min^{-1} at all the sites.

The concentrations of CO were measured continuously by using a nondispersive infrared analyzer (NDIR: GA-360S, Horiba, Ltd., Japan) with an overall analytical precision of $\pm 5 \text{ ppb}$. The measuring system has been changed to a gas chromatograph (TRA-1 Round Science Inc.), with an overall precision of $\pm 2 \text{ ppb}$, since 2008, 2009, and 2010 at YON, RYO, and MNM, respectively. The standard gases to calibrate the analyzers are traceable to the WMO mole fraction scale.

2.3 3-D chemical transport model

A 3-D chemical transport model (STAG) developed by AIST (Taguchi, 1996; Taguchi et al., 2002a) was used in this study. This model, which consists of 28 sigma

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vertical levels with a horizontal resolution of $1.875^\circ \times 1.875^\circ$, was driven every 6 h using the meteorological reanalysis data provided by the National Oceanic and Atmospheric Administration/the Cooperative Institute for Research in Environmental Sciences (NOAA/CIRES) Climate Diagnostics Center at their website (<http://www.cdc.noaa.gov/>). The boundary layer height at each time step in the model was determined by the bulk Richardson number (Troen and Mahrt, 1986) and a uniform distribution of trace gas concentrations was assumed in the boundary layer. The basic performance of the model was evaluated by using ^{222}Rn concentrations (Taguchi et al., 2002b).

The emanation rate of ^{222}Rn was given a value of $1.0 \text{ atom cm}^{-2} \text{ s}^{-1}$ for all the land areas between 60° S and 60° N , and $0.5 \text{ atom cm}^{-2} \text{ s}^{-1}$ for the land areas between 60° N and 70° N , excluding Greenland, with no temporal variation (Jacob et al., 1997; Taguchi et al., 2002b, 2011). The emanation rate of ^{222}Rn from the ocean was set to $0 \text{ atom cm}^{-2} \text{ s}^{-1}$, since our study focused on the short term variations of the atmospheric ^{222}Rn concentration.

CO emissions from all anthropogenic sources used in the model were obtained from the Emission Database for Global Atmospheric Research (EDGAR) version 4.1 (EC-JRC/PBL, 2010). Biomass burning, however, was not included. CO from photochemical production via the oxidation of CH_4 and nonmethane hydrocarbons (NMHCs) were regarded as constant with a value of 40 ppb, a value based on the result of previous simulation studies (Liang et al., 2004; Takigawa et al., 2005). Photochemical destruction of CO with hydroxyl (OH) radical was included in the model as the only CO sink. The rate coefficient of this photochemical destruction was obtained from Atkinson et al. (2006). The concentration of OH radical was obtained from the climatological seasonal fields published by Spivakovsky et al. (2000).

3 Results

3.1 Observational data of the atmospheric CO and ^{222}Rn

Figure 2 shows the hourly mean concentrations of atmospheric CO observed at MNM, YON, and RYO located in the Western North Pacific. All three stations reveal a distinct seasonal cycle accompanied by high-frequency synoptic variations, although the annual mean CO levels depend on the station. The seasonal cycle shows a minimum during the summer season from July to August and a maximum during the winter season from January to March. The synoptic variations consisting of episodic enhanced CO peaks on timescales less than one week are clearly revealed at all three stations, although their frequencies and magnitudes are different from each other. It has already been demonstrated by various modeling studies that major high CO concentration peaks observed at these stations are caused by the transport of Asian pollutions emitted mainly from China, Korea and Japan (Liang et al., 2004; Takigawa et al., 2005; Sawa et al., 2007).

Figure 3 shows the hourly mean concentrations of the atmospheric ^{222}Rn observed at MNM, YON, and RYO. The temporal variations of ^{222}Rn at all three stations show, as was the case with CO, a distinct seasonal cycle accompanied by high-frequency synoptic variations. No significant long-term trend was found at any of the stations. The difference in the magnitude of the ^{222}Rn concentration among the three stations is related to their respective geographical locations with different distances from the continental ^{222}Rn source regions. The seasonal cycles observed at MNM and YON are similar, with a maximum in the winter season around December–January and a minimum in the summer season around July–September. In contrast, a different pattern of seasonal cycle at RYO was found, reflecting a strong influence from local ^{222}Rn source emissions in a complex terrain surrounding the station.

No significant diurnal cycle was found at both MNM and YON, indicating no impact of local emissions on the ^{222}Rn measurements at these island stations. Thus, the numerous synoptic variations observed at MNM and YON were influenced by the long

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range transport of air masses from the Asian continent. In contrast, the distinct diurnal cycle was observed at RYO with a maximum in the early morning and a minimum in the afternoon. These were caused by the diurnal cycle in the venting rate associated with the planetary boundary layer (PBL) height over inland regions (Moses et al., 1960; Servant, 1966; Chambers et al., 2009). These results indicate a strong local influence on the ^{222}Rn measurements at RYO from the nighttime inversion.

3.2 Enhanced ^{222}Rn events

The observed concentrations of ^{222}Rn at three stations reveal numerous synoptic variations consisting of episodically enhanced ^{222}Rn peaks on timescales less than one week (Fig. 3). These peaks were caused by the transport of continental air masses since no local sources were found that would produce similar peaks, although RYO measurements are influenced by local sources. Based on the preliminary results obtained at MNM, these ^{222}Rn peaks showed high correlation with synoptic scale variation of CO observed simultaneously at this station (Wada et al., 2010).

Figure 4 shows the detailed variations of ^{222}Rn observed at MNM, YON, and RYO during January 2010, compared with those of observed concentrations of the atmospheric CO. During the month, 10, 9, and 4 enhanced ^{222}Rn events were identified at MNM, YON, and RYO, respectively. Corresponding increase in CO during each enhanced ^{222}Rn event is also shown in the figure. This ^{222}Rn -CO relationship was also observed at all three stations for other months during our study period. Each event corresponded to a cold frontal passage over the station, in a similar process described by Sawa et al. (2007) in which well-mixed polluted air masses with high concentration of CO from the Asian continent are transported to the Western North Pacific.

We identified the enhanced ^{222}Rn peaks following procedure described in Wada et al. (2011). The differences between the hourly observed data and the smoothed seasonal curve were calculated as residual ^{222}Rn (ΔRn), and are plotted in Fig. 4. Positive ΔRn peaks that were identified as enhanced ^{222}Rn (ERN) events were chosen based on the following criteria: (1) $\Delta\text{Rn} \geq 2\sigma$ from the fitted curve, where σ is the standard

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deviation calculated from the negative ΔRn in each month, and (2) ΔRn lasted for more than four hours. Following these criteria, 290, 516, and 148 ERN events were identified in the MNM data (September 2007–May 2011), the YON data (February 2006–May 2011), and the RYO data (March 2009–May 2011), respectively.

3.3 Correlation of observed CO with ^{222}Rn

In order to determine the enhancement ratios of CO and ^{222}Rn , slopes of ΔRn with ΔCO were calculated for each ERN event. ΔCO was determined by the same procedure as ΔRn . Figure 5 shows some examples of the enhanced CO correlations and their slopes for the ERN events observed at MNM, YON, and RYO in January 2010.

Since high correlations between CO and ^{222}Rn were found for the ERN events, their slopes were obtained by using the reduced major axis (RMA) regression (Hirsch and Gilroy, 1984). The slope of the enhanced values is denoted as $\Delta CO/\Delta Rn$. Note that the units of $\Delta CO/\Delta Rn$ are in $ppb (Bq m^{-3})^{-1}$. $\Delta CO/\Delta Rn$ obtained at MNM varied with each ERN event. These differences were found to be dependent on the transit time of the air masses from the Asian continent to MNM and the ^{222}Rn decay during the transport (Wada, 2011). The variation in $\Delta CO/\Delta Rn$ from ERN event to ERN event at MNM became negligible after correcting for the ^{222}Rn decay during transport. These results indicate that air masses reaching MNM from the continental Asia were well mixed by the time they reached the station. On the other hand, $\Delta CO/\Delta Rn$ at YON varied noticeably even though the ^{222}Rn decay correction was negligible due to short transit times of the air mass transport (Wada et al., 2011). The variation, therefore, is likely attributable to the variation in the sources themselves. In comparison, the ERN events at RYO were not that many and the ΔCO - ΔRn correlation was significantly low, due likely to the influence of local sources and complex terrain at RYO.

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3.4 Model simulation of the atmospheric ^{222}Rn and CO

^{222}Rn concentration was simulated by using the global chemical transport model STAG. Figure 6 shows a comparison of simulated ^{222}Rn concentration in the surface layer with the observed data of 6 hourly means at MNM, YON, and RYO for 2010. This model was generally successful in reproducing the seasonal variations observed at all three stations. The simulated ^{222}Rn in summer at MNM showed low values of less than 0.2 Bq m^{-3} , consistent with the observed concentrations. At YON, the observed ^{222}Rn values in the winter and spring seasons were well simulated by the model, although ^{222}Rn concentration in the model was slightly low when compared with the observation in the summer probably due to subgrid scale emissions of ^{222}Rn along the east coast of China. The simulated seasonal cycle at both YON and MNM agreed well with the observation. In this study, the model calculations were carried out by using a time invariant and spatially uniform ^{222}Rn flux density, although it was reported that ^{222}Rn fluxes over the continent were estimated to vary with time and space mainly due to changes in soil water content (Dörr and Munnich, 1990; Genthon and Armengaud, 1995). The consistency between the model and observation in this study indicated that the ^{222}Rn flux density employed in the model was sufficient for reproducing the observed ^{222}Rn concentration at these stations.

The model was also able to identify all the major enhanced ^{222}Rn in the synoptic scale variation observed at the stations, particularly in terms of the timing of the occurrences of these ERN events. However, the model slightly underestimated the magnitude of the ^{222}Rn enhancement, probably due to the coarse spatial resolution of the model.

The simulation of CO concentration was carried out by using the same model used for the simulation of ^{222}Rn . Figure 7 shows a comparison of simulated CO concentration in the surface air with the observed data of 6 hourly means at MNM, YON, and RYO for 2010. The model was successful in reproducing the observed seasonal cycle, synoptic variations, and enhanced CO peaks at these stations. Although the model

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underestimated the observed ^{222}Rn peaks, its underestimation of the observed CO peaks proved to be much more significant.

3.5 Relationship between observed data and simulated results for CO and ^{222}Rn

Figure 8 shows the relationships between the observed 6 hourly mean concentration and their simulated results for ^{222}Rn and CO for 2010 at the three stations. The correlation plots for ^{222}Rn show that the overall ratios of model/observation are about 0.72–1.00 with 95 % confidence interval of 0.02 calculated by the bootstrap method (e.g., Xiao et al., 2004), resulting mainly from the model underestimation of large ^{222}Rn peaks within subgrid scale variations. The data at MNM showed a compact relation with a high correlation coefficient of 0.84, while slightly low correlation coefficients of 0.64 and 0.47 were found at YON and RYO. Most of the major ERN events observed at all the stations throughout the year were captured by this model simulation. However, the correlation plots for CO at MNM and YON indicate that the model underestimated the observed CO, more significantly than for ^{222}Rn , due to the underestimated CO emissions in the EDGAR database that was used to drive the model CO field. This problem in the EDGAR database was also pointed out by previous modeling studies (e.g., Kiley et al., 2003; Arellano 2004; Ma and van Aardenne, 2004). Previous studies on the combustion ratio of CO/CO₂ observed around Western North Pacific indicated that YON is influenced mostly by the emissions from China where anthropogenic emissions have been significantly underestimated (e.g., Palmer et al., 2003; Heald et al., 2004). On the other hand, RYO is influenced mostly by regional sources in Japan, while MNM is influenced by sources distributed over a much greater areas in East Asia (Wada et al., 2011). Therefore the emission of CO at the stations of MNM and YON are significantly underestimated when compared with that of RYO.

Among all the stations, MNM is located farthest from the continental East Asia. It therefore “sees” CO emissions from a wider geographical region in East Asia than the other stations. Combined with the fact that CO and ^{222}Rn are highly correlated at MNM,

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we used the observed and simulated data at MNM to obtain a more realistic estimate of CO emission from East Asia than before.

3.6 Estimation procedure based on the radon tracer method

The radon tracer method has been mainly applied to inland sites in European. We changed the method to apply to our sites in Western North Pacific region. The basic concept of our method is that the enhancement ratio of CO/²²²Rn associated with each ERN event show agreement between the model and observation when the emission database used in the model are realistic. Error in either CO or ²²²Rn emission in the model can produce disagreement with the observed CO/²²²Rn ratio. In our study, the model simulated the observed ²²²Rn well. Therefore, any difference between the modeled and the observed CO/²²²Rn ratio can be traced to error in the estimated CO emission in the model, which can be adjusted to bring the model simulation results closer to the observed. This way, we were able to obtain a more realistic estimate of CO emission from East Asia by using the CO/²²²Rn ratio. It should be noted that the uncertainty in the model transport was viewed as not a significant issue because the same transport model was used to simulate both CO and ²²²Rn.

We conducted a tagged tracer experiment to adjust the simulated CO so that the simulated CO/²²²Rn ratio would match the observed ratio. The adjustment is expressed as a cost function by which the CO emission is optimized (e.g., Suntharalingam et al., 2004; Sawa et al., 2007). Figure 9 shows the regions used for the tagged tracer experiment. The global emission area is divided into 14 regions. China was divided into 3 regions of North China (NC), South China (SC), and West China (WC), with their boundaries defined by 100° E longitude and 35° N latitude. Japan and Korea were combined into one region (JpK).

The adjusted simulated CO was obtained by the following equation;

$$X'_{\text{CO}} = \sum_{i=1}^n k_i C_i. \quad (1)$$

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where X'_{CO} represents the adjusted simulated CO at MNM, n is the number of regions (equal to 14 in our study), C_i is the simulated CO at MNM with influence from region i obtained by the tagged tracer experiment, and k_i is the corrective coefficient for region i . These corrective coefficients were determined by minimizing the cost function which was defined in this study as residual sum of squares between the observed and the simulated ratios associated with each ERN event. The cost function is described by the following equation;

$$S = \sum_{j=1}^m \left(\left(\frac{X_{CO}}{222Rn} \right)_j^{\text{observed}} - \left(\frac{X'_{CO}}{222Rn} \right)_j^{\text{simulated}} \right)^2. \quad (2)$$

Where S is the residual sum of squares and m is the number of ERN events. Through changing the corrective coefficients of k_i in Eq. (1), optimized pairs of k_i are obtained by minimizing S in the Eq. (2).

3.7 Estimates of CO emission in East Asia

Figure 10 shows the results of the tagged tracer experiment for CO. The shaded areas represent simulated ERN events for the peaks 1, 4, 7, 8, 9, and 10 of the observed ERN events shown in Fig. 4. Other peaks were not well reproduced because of the relative smallness of the events. Figure 10 shows that the synoptic scale variation of atmospheric CO at MNM was influenced by three regions of NC, SC, and JpK, while other regions were negligible. CO emission from the biomass burning in Southeast Asia was also deemed negligible based on the results of Sawa et al. (2007) who conducted a similar tagged tracer experiment for CO in March using the same transport model with temporally variable biomass burning emission. Consequently, the corrective coefficients in Eq. (1) for the regions NC, SC and JpK were adjusted to optimize the CO emissions in the model, whereas the corrective coefficients for other 11 regions were fixed to be 1.

To perform calculations using Eq. (2), the ERN events observed from November to April were selected in order to ensure cold-front transport mechanism for all the events. In addition, the ERN events were characterized by high correlation between enhanced CO and ^{222}Rn for both the observed and simulated results, ensuring pollution events with well mixed air masses. Twenty ERN events with correlation coefficient of more than 0.7 were found, constituting about 11.2 % of the identified ERN events during November to April. With these 20 ERN events, the corrective coefficients for the regions of NC, SC, and JpK were determined by changing each corrective coefficient from 0.5 to 3.0. From the result of the optimization, the corrective coefficients for NC, SC, and JpK were determined to be 2.8, 1.4, and 1.1, respectively. Figure 11 shows an example of the correlation between CO and ^{222}Rn for the ERN event on 23 November in 2009 from the observation and simulations. In this figure, a priori ratio of CO/ ^{222}Rn calculated by the model was significantly smaller than the observed ratio due to the aforementioned underestimation of CO emission in East Asia, whereas a posteriori ratio improved after adjusting the CO emissions in the 3 regions. The adjustment resulted in a new emission estimate of 203 TgCOyr⁻¹ for China, Japan, and Korea, with 185 TgCOyr⁻¹ attributed to China. Although the criterion to select an ERN event was based arbitrarily on correlation coefficient being greater than 0.7, changing the criterion from 0.6 to 0.8 resulted in one standard deviation variability of only 8.3 % in emission estimates.

3.8 Comparison of CO emission estimates

Table 1 shows a comparison of the CO emission for East Asia obtained in this study with emission estimates from several inversion and bottom-up methods. Our study shows that 93 % of the total emission for East Asia comes from China. The remaining comes from Japan and Korea. This is consistent with the results of Palmer et al. (2003), Kopacz et al. (2009), and three bottom-up emissions. There are however, large variations in the emission estimates within each method, as well as between the methods. Among the bottom-up methods, the EDGAR database shows the greatest underestimation. The variation among the inverse methods could be attributable to an increase

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in the emission during 2001 to 2005. Tanimoto et al. (2008) reported that an increase in the CO emission in China from 2001 to 2005 was estimated to be about 16 %. We used this rate of increase to adjust all the emission estimates listed in Table 1 to a reference year of 2005. Figure 12 shows the estimated emissions of CO in China listed in Table 1 (solid bars) and their adjusted values to the reference year 2005 (open bars). From the figure, it can be seen that the EDGAR database shows a significant underestimation by about 45 %. All other estimates, including the one from this study, agree reasonably with each other. Ohara et al. (2007) reported that the REAS database was consistent with the inverse estimates and those from the forward modeling, whereas Zhang et al. (2009) reported that the INTEX-B database was partially consistent with a forward modeling result (Chen et al., 2009) at a suburban area near Beijing but underestimated in the Beijing urban area.

4 Conclusions

Atmospheric concentrations of CO and ^{222}Rn were observed continuously at Minamitorishima (MNM), Yonagunijima (YON), and Ryori (RYO) from 2007 to 2011. These measurements showed a seasonal cycle with a winter maximum and a summer minimum in both CO and ^{222}Rn . In addition, high-frequency synoptic variations associated with enhanced ^{222}Rn (ERN) events due to a long-range transport of continental air masses were well captured.

The enhancements of CO associated with increased ^{222}Rn were found in most of the ERN events at MNM, YON, and RYO. These simultaneous elevations indicate that the concentrations of ^{222}Rn and CO were well mixed in the pollutant air masses over the continent and were exported to the Western North Pacific region by the way of cold frontal transport. The correlation of ^{222}Rn and CO during each ERN event observed at MNM was very high, with very little variation in the CO/ ^{222}Rn ratio from event to event during winter. The correlation of the enhancement ratio at YON was also high during winter and spring, but the ratio varied from event to event due the station's vicinity to

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source regions, whereas the ratio at RYO was significantly low due to influence from local sources in a complex terrain.

The chemical transport model STAG reproduced well the concentration of observed ^{222}Rn when forced with constant and uniform flux density of $1.0 \text{ atom cm}^{-2} \text{ s}^{-1}$. The simulated seasonal cycles and synoptic scale variations were in good agreement with the observed. The model also reproduced well the timing of the synoptic scale variation of the observed CO, but underestimated the magnitude of the variation. The ratios of the simulation/observation of CO were 0.52 and 0.42 at MNM and YON, respectively, whereas those of ^{222}Rn were 0.72 and 0.81. The relatively low ratio values resulted from using in the model the EDGAR emission database that is known to have a significant underestimation of CO emission values over China.

The tagged tracer experiment of CO at MNM indicated that the synoptic variation of CO at MNM was influenced by the emissions from North China (NC), South China (SC), and Japan and Korea (JpK), with emissions from other regions found to be negligible in the winter and spring.

An estimation procedure based on the radon tracer method was developed and applied to the measurements from MNM. By optimizing the a priori emission of the EDGAR ver. 4.1 database for the ERN events observed at MNM during the winter and spring, the total anthropogenic CO emission from China, Japan, and Korea was estimated to be $203 \text{ Tg CO yr}^{-1}$, of which 93 % originated in China. When compared with other estimated emissions of CO based on top-down and bottom-up methods, our estimated result was found to be in general agreement, except for the emission database of EDGAR ver. 4.1 which came out to be about 45 % smaller than our emission estimate for China. These results strongly indicate that an independent method, such as the radon tracer method used in this study, to estimate emissions is important in East Asia where there is still large uncertainty associated with various estimates.

Acknowledgements. We would like to acknowledge all members of the Japan Meteorological Agency for operating ^{222}Rn and CO measuring systems at the stations of Minamitorishima, Yonagunijima, and Ryori. We also thank T. Ohara and J. Kurokawa for providing emission data

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base of REAS 2005. We wish to thank K. Higuchi for useful comments and discussion. Our acknowledgements are extended to A. Kudo and C. Abe for maintenance of the ²²²Rn measuring systems.

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Table 1. Comparison of estimated CO emissions in East Asia.

Method	Study	Sources**	Tg CO yr ⁻¹	Regions	Reference year
Our Method	This work	FF; BF	203	China, Japan, Korea China	2007–2011
			185		
Inverse Method	Tanimoto et al. (2008)	FF; BF; BB	170	China	2005
	Heald et al. (2004)	FF; BF; BB	192	China, Japan, Korea	2001
	Palmer et al. (2003)	FF; BF	194	China, Japan, Korea China	2001
			168		
	Wang et al. (2004)	FF; BF; BB	170	China	2001
	Kopacz et al. (2009)	FF; BF; BB	156	China, Japan, Korea China	2001
			142		
Yumimoto and Uno (2006)	FF; BF	147	China	2001	
Arellano (2004)	FF; BF	205	China, Japan, Korea	2000	
Bottom-Up Emission	EDGAR v4.1	FF; BF	118*	China, Japan, Korea China	2005
			102*		
	REAS	FF; BF	206*	China, Japan, Korea China	2005
			192*		
INTEX-B	FF; BF	170*	China, Japan, Korea China	2006	
		159*			

* These values were summed in each region defined in this study.

** FF – fossil fuel; BF – Biofuel combustion, BB – biomass burning

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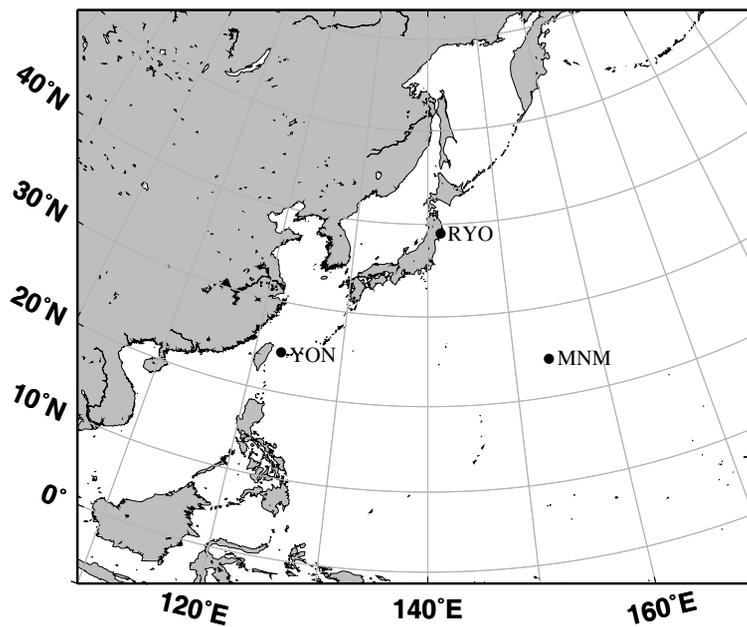


Fig. 1. Geographical locations of Minamitorishima (MNM), Yonagunijima (YON), and Ryori (RYO).

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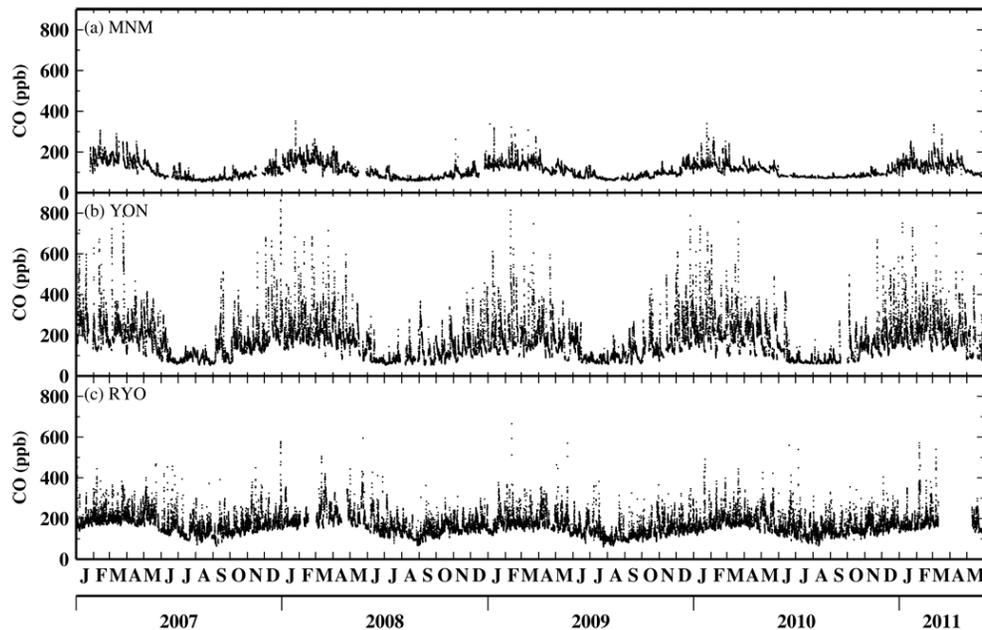


Fig. 2. Hourly mean observed data of the atmospheric CO concentration at MNM (a), YON (b), and RYO (c).

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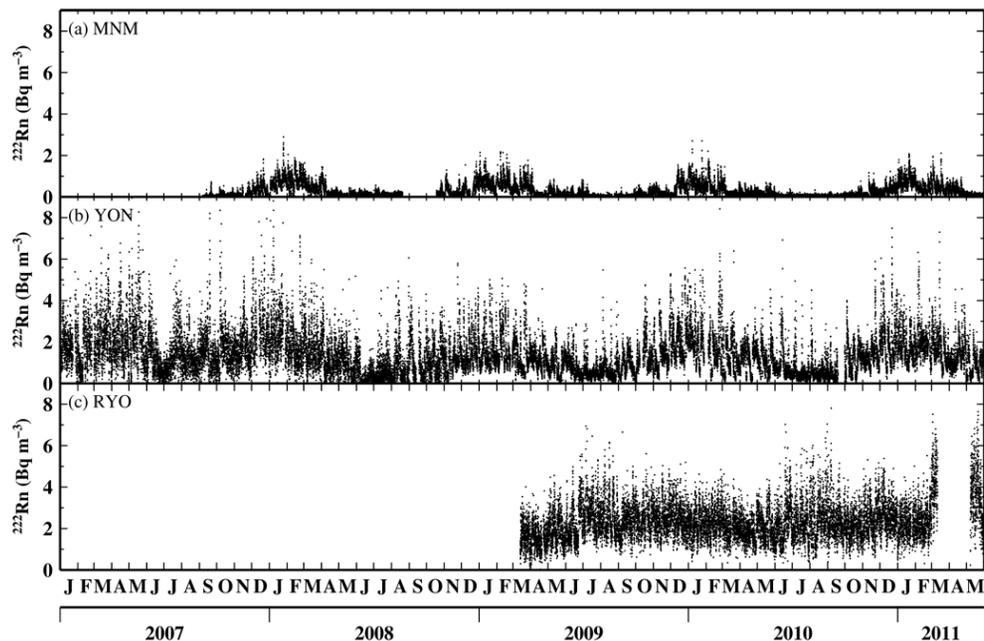


Fig. 3. Hourly mean observed data of the atmospheric ^{222}Rn concentration at MNM (a), YON (b), and RYO (c).

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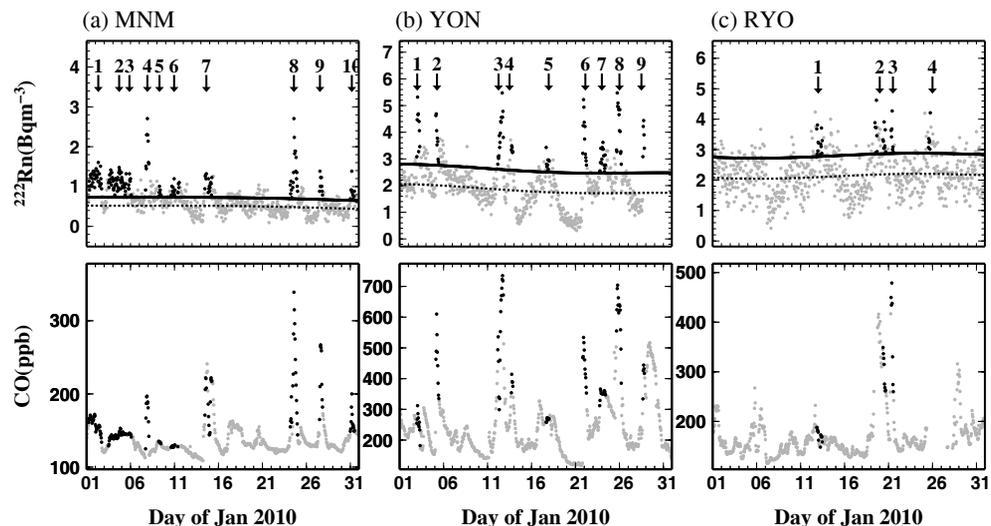


Fig. 4. Typical synoptic scale variation of hourly mean concentrations of atmospheric ^{222}Rn and CO observed at MNM (a), YON (b), and RYO (c) in January 2010. Arrows represent the peak of each enhanced ^{222}Rn event. The dashed and the solid lines for ^{222}Rn (top panel) are the smoothed fitting curve and the upper threshold of ΔRn , respectively. The black and gray dots represent the ΔRn data defined as enhanced and non-enhanced events, respectively (see text). The black dots for CO relate to the corresponding periods of the enhanced ^{222}Rn events.

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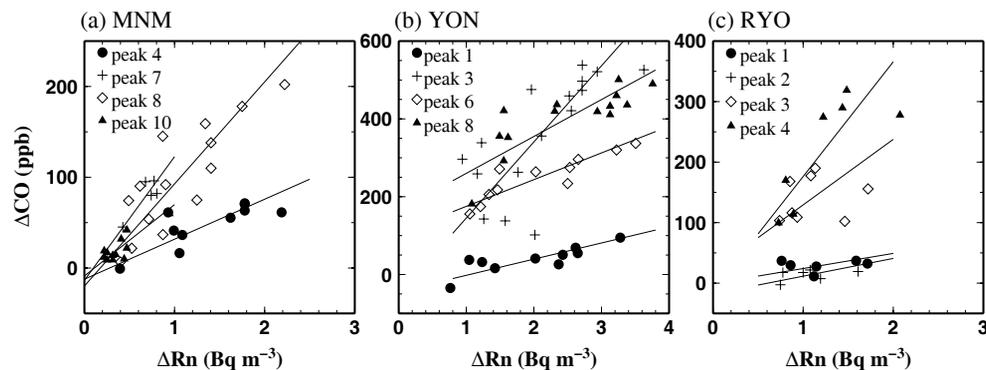


Fig. 5. Typical scatter plots of ΔCO as a function of ΔRn observed at MNM (a), YON (b), and RYO (c) in January 2010. The solid line represents the RMA regression for each event. The peak numbers correspond to those in Fig. 4.

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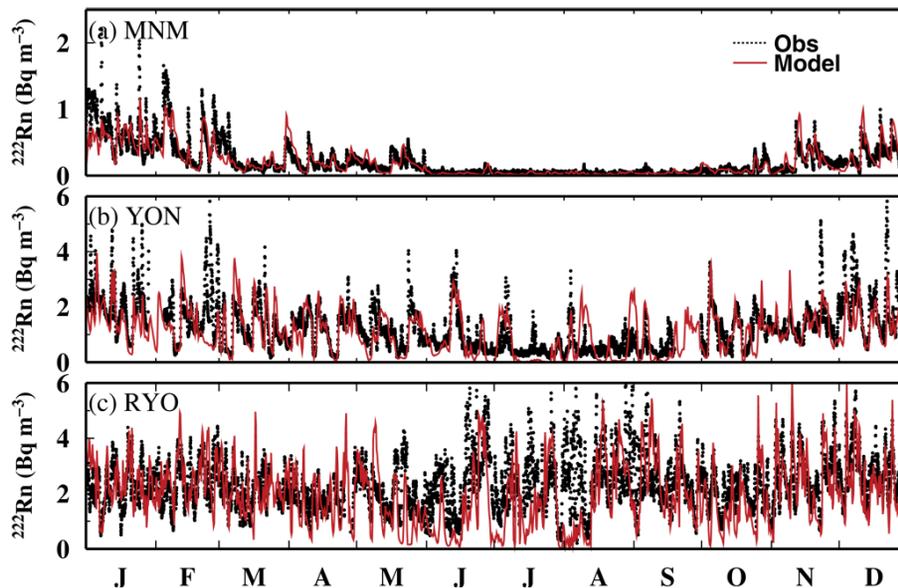


Fig. 6. Simulated and observed atmospheric concentration of ^{222}Rn at MNM (a), YON (b), and RYO (c) in 2010. The black dots represent 6 hourly mean observed ^{222}Rn and the red solid line represents simulated ^{222}Rn .

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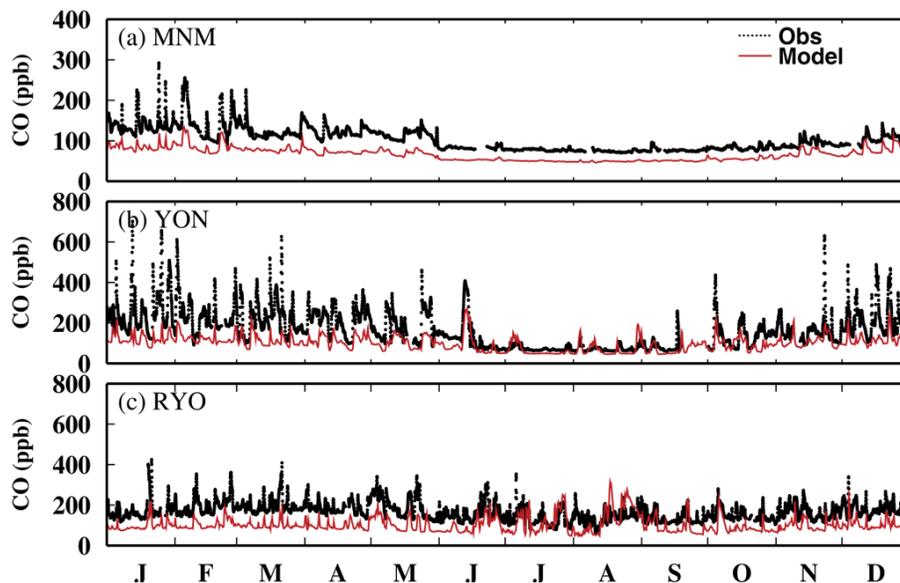


Fig. 7. Simulated and observed atmospheric concentration of CO at MNM **(a)**, YON **(b)**, and RYO **(c)** in 2010. The black dots represent 6 hourly mean observed CO and the red solid line represents simulated CO.

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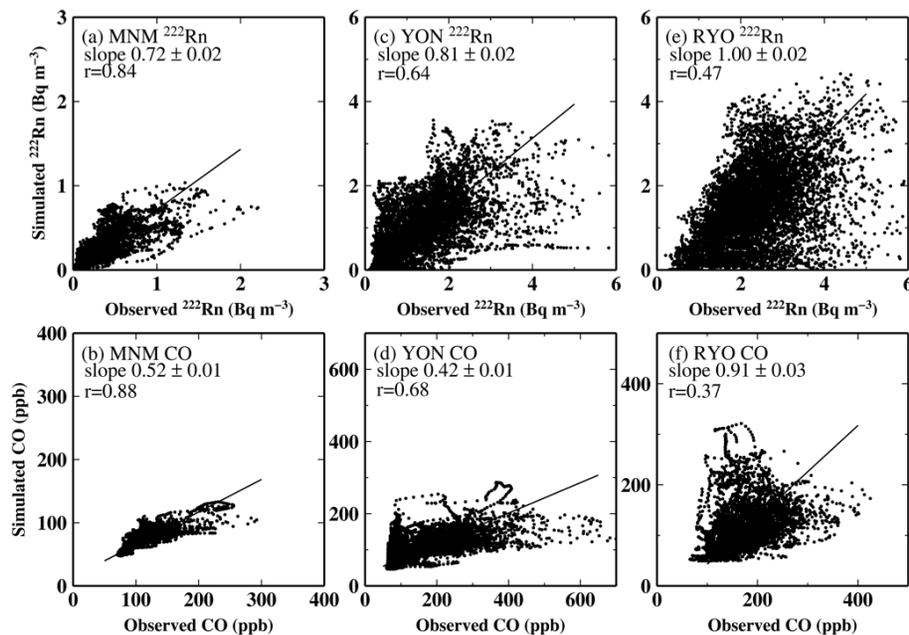


Fig. 8. Correlations between simulated results and observed data for ^{222}Rn and CO in 2010 for MNM, YON, and RYO. The solid lines represent the RMA regressions. The slopes are calculated by the RMA regression with 95 % confidence interval obtained by the bootstrap method. The r values represent correlation coefficients.

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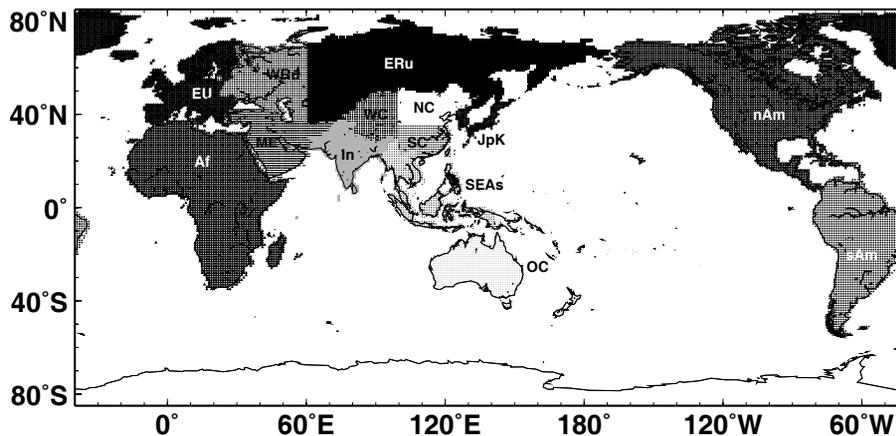


Fig. 9. Regions divided into 14 sectors for the tagged tracer model experiment. JpK; Japan and Korea, NC; North China, SC; South China, WC; West China, SEAs; Southeast Asia, In; India, OC; Oceania, ERu; East Russia, WRu; West Russia, ME; Middle East, EU; Europe, Af; Africa, nAm; North America, sAm; South America.

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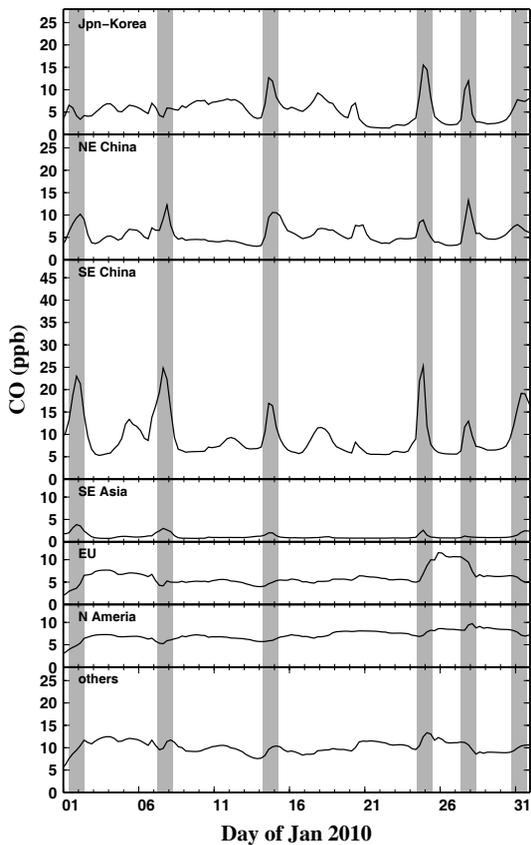


Fig. 10. Simulated results of the tagged tracer experiment for atmospheric CO at MNM. The gray shaded areas represent the enhanced CO events for peak 1, 4, 7, 8, 9, and 10 of the observed data in Fig. 4.

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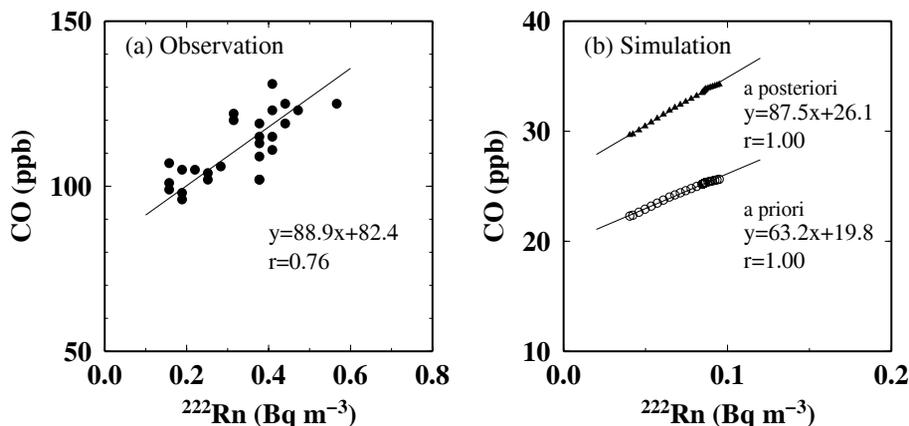


Fig. 11. An example of correlations between CO and ^{222}Rn for the enhanced ^{222}Rn event on 23 November in 2009 from the observation (a) and simulations (b) with a priori (open circles) and a posteriori (solid triangles) emissions, respectively. The solid lines represent the RMA regressions and the r values represent correlation coefficients.

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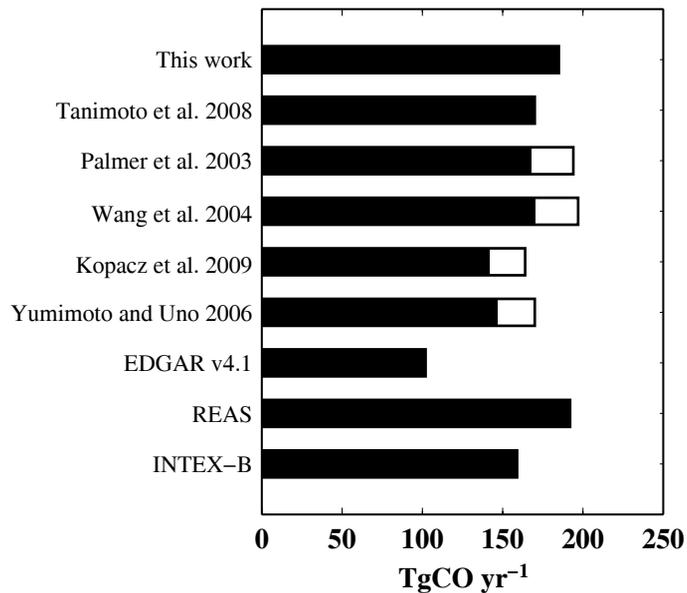


Fig. 12. The estimated emission of CO in China. The solid bars represent data in Table 1, and the open bars represent estimated values for the reference year of 2005 by assuming an increase of 16 % during 2001–2005.

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