Chapter 6
Continuous Monitoring and the Source Identification of Carbon Dioxide at Three Sites in Northeast Asia During 2004–2005

Fenji Jin[1], Sungki Jung[1], Jooll Kim[1], K.-R. Kim[1], T. Chen[2], Donghao Li[2], Y.-A. Piao[2], Y.-Y. Fang[2], Q.-F. Yin[3], and Donkoo Lee[4]

Abstract We conducted continuous monitoring and the source identification of carbon dioxide at Gosan, Seoul (Korea) and Yanbian during 2004–2005. The data reported are in situ continuous 1-year measurements of atmospheric CO_2 from the Gosan, Seoul and Yanbian stations. One-minute averages of near-surface atmospheric CO_2 concentration were obtained using a measurement system based on non-dispersive infrared (NDIR) analysis using the NOAA/ESRL (National Oceanic & Atmospheric Administration/Earth System Research Laboratory) standard with high precision monitoring data. The background CO_2 concentration of the complete measurement data was determined using the Advanced Global Atmospheric Gases Experiment (AGAGE) statistical pollution identification procedure for removing pollution episode data. The background characteristics at Gosan are discussed in detail. The background concentration of CO_2 showed quite evident diurnal and seasonal variation. The diurnal variation shows a maximum in the nighttime and a minimum in the daytime, and the seasonal cycle shows a maximum in spring and a minimum in summer. Background data at Seoul and Yanbian also show a similar trend. In addition, we applied a hybrid receptor model driven by three-dimensional synoptic meteorology from the HYSPLIT4 (HYbrid Single-Particle Lagrangian Integrated Trajectory) model to determine CO_2 relative emission strength contributions from the Northeast Asia region as observed from Gosan. Modeling results from Seoul and Yanbian are also presented—they are important in creating a full potential source region map of the Northeast Asia region, as observations from Gosan are limited to the wind patterns crossing the station. Results indicate that there appears to be a large potential source region in the northeastern and eastern parts of China.

Keywords: Carbon dioxide, continuous monitoring, emission strength, long-range transport, trajectory

[1] *School of Earth and Environmental Science, Seoul National University, Seoul, Korea*

[2] *Yanbian University, Yanji, Jilin, China*

[3] *Huaiyin Teacher's College, Huaian, Jiangsu, China*

[4] *College of Agriculture and Life Sciences, Seoul National University, Seoul, Korea*

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6.1 Introduction

The role of greenhouse gases derived human activities in the radiative forcing of climate change has been well documented in the scientific reports of the Intergovernmental Panel on Climate Change (IPCC 2001). Carbon dioxide (CO_2) is one of the representative greenhouse gases. Before the Industrial Era, which began around 1750, the atmospheric carbon dioxide concentration had remained constant at 280 ± 10 ppmv for several thousand years. It has risen continuously since then, reaching 380 ppmv in 2004. The continuous rise in levels of atmospheric CO_2 is caused by anthropogenic emissions of CO_2. In particular, three-quarters of these emissions are due to fossil fuel burning (IPCC 2001).

Continuous observation of CO_2 mixing ratios in the atmosphere is very important, providing a basis for studies of the global carbon cycle and CO_2-induced climatic change. For this reason atmospheric CO_2 has been monitored at many sites worldwide for many years. Northeast Asia is of special interest because the economic growth of this region during the last 50 years could have significant implications in terms of the release of industrial pollution into the environment, including anthropogenic release of CO_2 into the atmosphere. Although measurement data obtained through frequent flask sampling can show seasonal variations, it is difficult to determine detailed variations such as diurnal variation and CO_2 concentrations affected by air mass transport on an hourly timescale. With the goal of establishing a background reference site for future research, the Gosan station at Jeju Island, Korea was created in 2003.

Deriving the background concentration from the continuous atmospheric measurement of a specific species is an important objective for which many different methods have been developed, such as the weighted method (Gras 2001; Zhou 2003), the European Monitoring and Evaluation Program (EMEP) daily wind direction sector allocations, the Nuclear accident model (NAME), the Lagrangian dispersion model defined air mass origins, and the AGAGE statistical pollution identification procedure (O'Doherty et al. 2001). For this study we have utilized the AGAGE statistical method of background calculation (O'Doherty et al. 2001).

An important application of measurement data is the modeling of the emission source and the magnitude of emissions from the surrounding region. Air-mass back trajectories have often been used in combination with observational data to identify potential source areas of air pollutants and determine their respective contribution at the receptor sites. Many statistical methods exist for this purpose, including residence time analysis, and hybrid receptor models such as PSCF (Potential source contribution function), CWT (concentration weighted trajectory), and RTWC (residence time weighted concentration) (Ashbaugh 1983; S. Reimann 2004). Here, we apply the RTWC model of back trajectory calculated in consecutive time steps, 24 times a day and CO_2 concentration in the same steps. Following the example of Gosan station, we have gradually expanded our *station* to other sites, namely Yanbian (China) and Seoul (Korea) to extend the potential source region identified.

The objectives of this study are source identification of the carbon dioxide in the Northeast Asia region and model calculations of relative emission strengths in these regions through in-situ continuous monitoring measurement at Gosan, Seoul and Yanbian. These results may have significant implications, as accurate CO_2-emission data is becoming important with efforts to reduce global atmospheric pollution such as the Kyoto protocol.

6.2 Methodology

6.2.1 Site Description

Figure 6.1 shows a map of the Northeast Asia region with three of the measuring stations used in this study. A detailed description follows.

The Gosan station is located near the southwestern tip of the Jeju Island, south of the Korean peninsula (126°11′00″ E, 33°11′70″ N; altitude: 70 m above mean sea level). Because of its remote and relatively unpolluted location, Gosan station is considered to be an ideal location for measuring the atmospheric composition of air masses considered to be representative of the background concentrations in Northeast Asia (Carmichael et al. 1997; Chen et al. 1997). For this reason, Gosan station has been included in numerous cooperative research projects, like ACE-Asia and Atmospheric Brown Cloud, as well as being incorporated into research networks such as the CSIRO-LOFLO network and the Advanced Global Atmospheric Gases Experiment (Prinn et al. 2000) networks. We have conducted high-frequency continuous monitoring at Gosan, Jeju, since 2003.

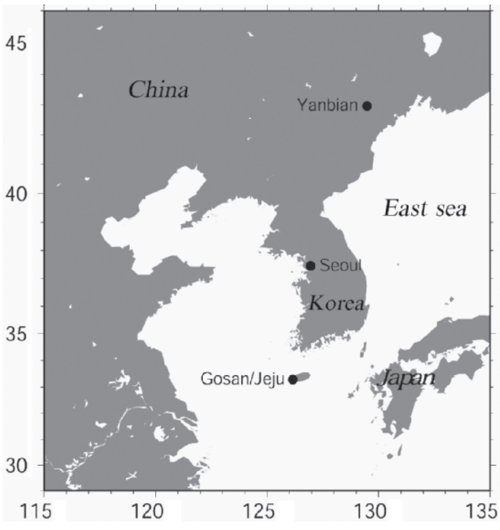

Fig. 6.1 Atmospheric monitoring sites showing Gosan/Jeju island station, Seoul station and Yanbian station, respectively

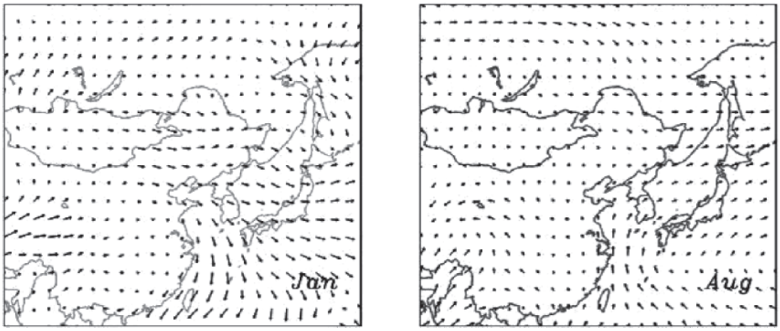

Fig. 6.2 Monthly wind for the Northeast Asia region from NCEP (2005)

Although too close to local pollution sources to be valuable as background monitoring stations, Seoul and Yanbian stations could still be used to observe regional pollution and thus be utilized for broadening the potential source region study area. Seoul station (37°27′N, 126°57′E) is located in Seoul National University, and is considered representative of atmospheric CO_2 mixing ratios in a highly industrialized city. Measurements from Seoul will prove to be valuable in determining the baseline of the Korean peninsula as well as research into pollution sources in Korea and China. Continuous measurements have been conducted there since 2005. Yanbian station (42°32′ N, 129°18′ E) is located at Yanbian University, China. Yanbian city is a semi-developed city with a population of just over two million. The relatively less developed nature of its surroundings could be ideal for studying the air mass outflow from the Asian continent, with an emphasis on pollution from China. Continuous measurements of atmospheric CO_2 have been conducted there since 2004.

Analysis of the monthly wind provided by the US National Centers for Environmental Prediction (NCEP) global data assimilation system (GDAS) for the Northeast Asia region shows that the prevailing winds in the region are northwesterly in winter and southwesterly in summer (Fig. 6.2).

6.2.2 Analytical Method

Atmospheric CO_2 concentration is measured using a Licor 6262 non-dispersive infrared (NDIR) analyzer system (Fig. 6.3). The system is composed of four parts, inlet part, controller part, calibration part and detector part. Ambient air is supplied into the system via 10 mm o.d. Dekoron tubing 40 m up to the intake tower. Air is drawn in via a vacuum pump through pressure release valves set at 6 psi to remove excess air and allow high continuous flow through the mainline and passed through a 7 µm in-line filter to remove particles and a Nafion-dryer to remove moisture.

NOAA/ESRL (National Oceanic & Atmospheric Administration/Earth System Research Laboratory) reference standards were used as the calibration reference, with

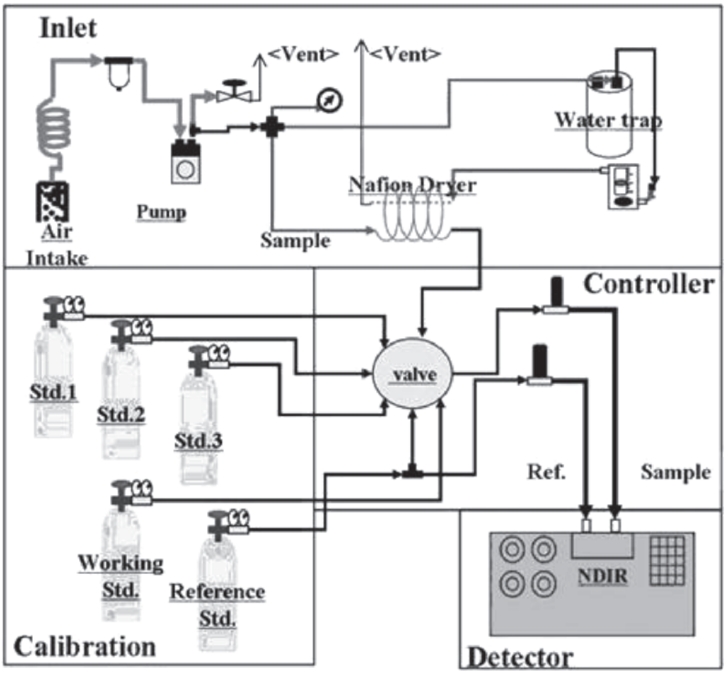

Fig. 6.3 Continuous CO_2 monitoring system diagram

3 standards per station in the range of 300–400 ppmv. The sample precision through an NDIR analyzer was below ±0.1 ppmv. The NDIR system was installed at each of the stations; at Gosan in April 2004, at Yanbian in January 2005 and at Seoul in March 2005. Air samples were analyzed at a frequency of two to three times per minute.

6.2.3 Background Determination

In previous studies different methods were used to sort the observations by air mass origins, in an attempt to separate regional and/or local pollution events from the background measurement (Gras 2001; Zhou 2003). Because air masses from various sources are influencing the in-situ site, the approach of selecting a certain wind sector as the background condition requires careful measurement and analysis of the wind patterns of the monitoring location, and is not easy to implement in all locations. So in the present study the AGAGE statistical pollution identification procedure was selected to determine the background concentration (O'Doherty et al. 2001).

AGAGE statistical pollution identification procedure determines the pollution events of a given day by examining the trends for 60 days before and after it.

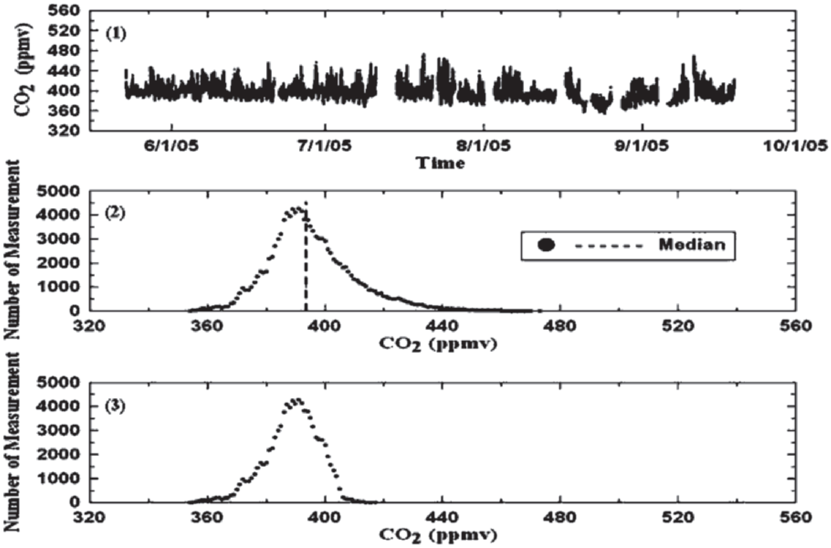

Fig. 6.4 Pollution identification for Seoul, measurements, (**1**) plot of 121-day window around selected measurement, (**2**) median (hashed line) values of all sample points, (**3**) remaining baseline data with Gaussian distribution

A Gaussian distribution is assumed for the background during this 121-day period, and events that deviate from the median of the distribution by more than a certain factor (typically 2–3 σ) are labeled as pollution events. Details of the method are explained in the Appendix of O'Doherty's paper (O'Doherty et al. 2001).

Figure 6.4 depicts the validity of the AGAGE statistical pollution identification process. Figure 6.4(1) shows the time series with identified pollution events of CO_2 from May 18 and September 18, 2005 at Seoul. Figure 6.4(2) shows that the histogram indicates that the majority of CO_2 data during this period is centered about 390 ppmv and the long tail feature of high CO_2 concentration values are less frequent. After applying the AGAGE statistical pollution identification procedure, the remaining data (background data) show a structure similar to a Gaussian distribution (Fig. 6.4(3)).

6.2.4 Hybrid Receptor Model

From April 2004 to March 2005, three-day back trajectories for every hour were calculated by the HYbrid Single-Particle Lagrangian Integrated Trajectory (HYSPLIT) model (Draxler and Rolph 2003) with 6-hourly archived meteorological data provided from the US National Centers for Environmental

Prediction (NCEP) global data assimilation system (GDAS), known as the final run (FNL) data.

In order to investigate potential source regions of CO_2 we combined these trajectories with measured concentrations at the station. For this we have used the method of Seibert et al. (1994) which computes the mean concentration for each grid cell after superimposing a grid to the domain of trajectory computations using the following formula:

$$\overline{C_{ij}} = \frac{1}{\sum_{l=1}^{M} \tau_{ijl}} \sum_{l=1}^{M} (c_l) \tau_{ijl},$$

where $\overline{C_{ij}}$ is a relative measure of potential source region strength, i and j are the indices of the horizontal grid, l is the index of the trajectory, M is the total number of trajectories, c_l is the concentration (minus the background concentration) measured during arrival of trajectory l, and τ_{ijl} is the residence time of the trajectory l spent over grid cell i, j. This concentration field method was used for several compounds to investigate their source (Stohl 1996; Charron et al., 2000; Ferrarese et al., 2002; Reimann et al., 2005). The domain of the calculated trajectories was superimposed with a 0.5° × 0.5° grid. For the calculation of residence time, we used the method of Poirot et al. (1986) with adjustments applied for geometry.

A high value of $\overline{C_{ij}}$ means that, on average, air parcels passing over cell (i,j) result in high concentrations at the receptor site. But because measured concentrations are distributed equally to all grid cells passed by the appropriate trajectory, the approach used is susceptible to underestimation of the spatial gradients of the true emission field (Stohl 1996). In order to eliminate low confidence level areas, a point filter was applied to the model results. This increases the confidence level of the results but also reduces the area studied.

6.3 Results and Discussion

6.3.1 Data

Plots of the continuous monitoring data for each of the stations are shown in Fig. 6.5.

The gray points represent the pollution flagged data using the statistical pollution detection algorithm described in section 6.2.3, and the black points are the remaining background data. The frequency and amplitude of the pollution events at Yanbian and Seoul are noticeably larger, due to the urban characteristics of their locations. A more detailed discussion of the background data follows, and the pollution data will be discussed in the next section.

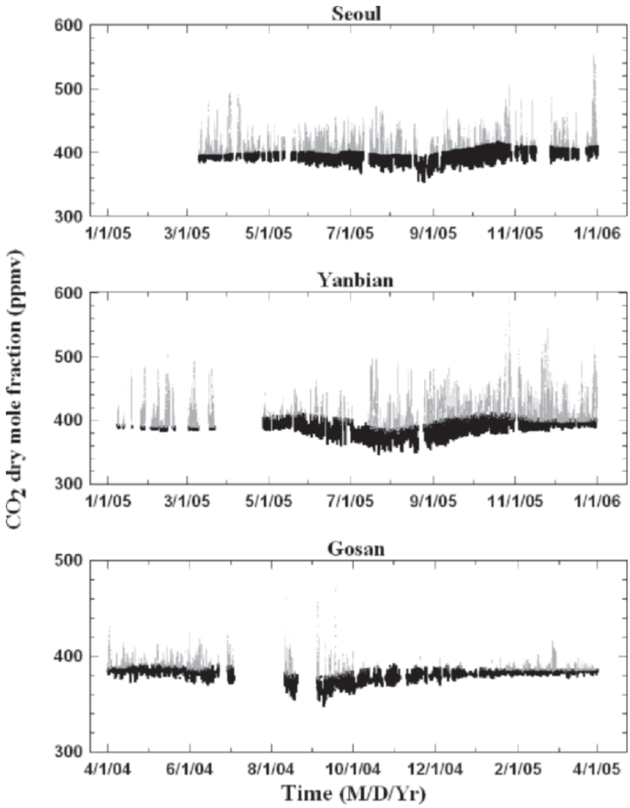

Fig. 6.5 Continuous monitoring (1 min) CO_2 data measured at each of the Northeast Asia Atmospheric Monitoring Network sites. Time shown is the local time

6.3.2 Atmospheric CO_2 Background Characteristics

Applying the AGAGE statistical pollution identification procedure we derived the background CO_2 concentrations in Gosan, Seoul and Yanbian.

6.3.2.1 Diurnal Variation

Background concentrations of CO_2 at Seoul, Yanbian and Gosan show diurnal variation (Fig. 6.6), higher values in the nighttime and lower values in the daytime. The diurnal variation is relatively small in winter and large in summer, due to the effect of photosynthesis by plants being stronger in daytime and summer than in nighttime and winter.

All of the stations show higher concentrations in the nighttime and lower concentrations in the daytime. The high-peak at Gosan appears at approximately 6 h

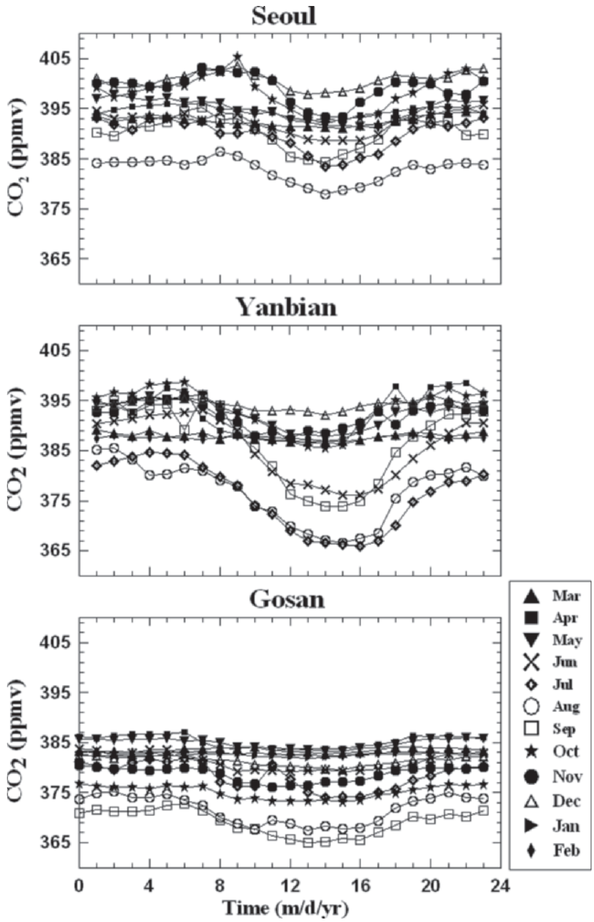

Fig. 6.6 Averaged CO_2 diurnal variation of background data at Seoul, Yanbian and Gosan in the measurement period. Hour in the shows the local time in each site

and the low peak around 13–15 h. The high-peak for Yanbian is at around 6–8 h, with the low-peak at 14–16 h. Seoul shows a high-peak at 7–9 h and a low-peak at 13–15 h. All of the times are in local time. One reason for this trend is the respiration in nighttime and photosynthesis in daytime by plants. Another reason is the change in the boundary layer, which changes with the earth's surface temperature. This change will enhance the low concentrations in the afternoon and the high concentrations in the early morning.

The amplitude of the diurnal variation is largest in summer, due to increased influence from plants. The smallest diurnal variation can be seen in Gosan, with Yanbian showing the largest variations. This can be attributed to the relatively low influence of terrestrial plants in Gosan, while Yanbian station is more influenced by terrestrial plants due to the surrounding forests.

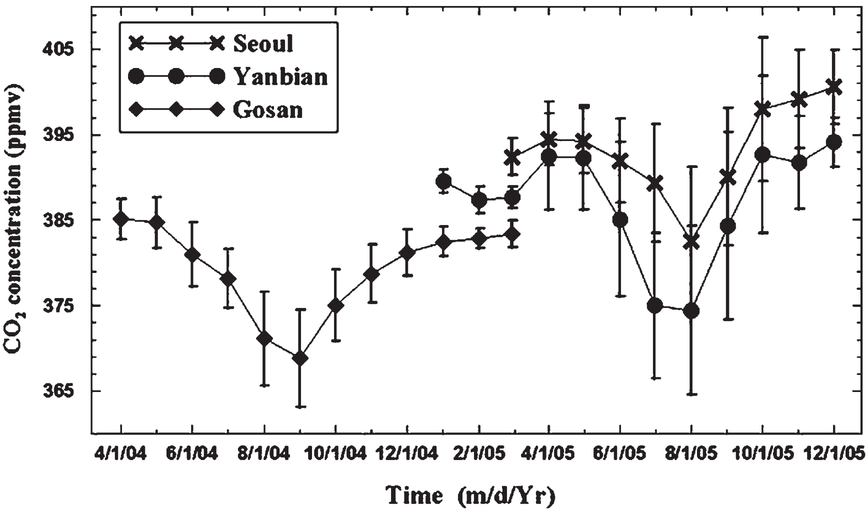

Fig. 6.7 CO_2 averaged seasonal cycle of background data at Seoul, Yanbian and Gosan with standard deviation indicated as error bars in the measurement period

6.3.2.2 Seasonal Variation

The averaged CO_2 seasonal variation for the background data is shown in Fig. 6.7.

There was an obvious seasonal cycle, with a maximum occurring in April and a minimum in September at Gosan. The measurements from Gosan show a low peak at September, but not having a large enough number of measurements in August may have caused this phenomenon. Future measurements will verify this anomaly. CO_2 mixing ratios declined rapidly during the period May–September, and climbed fleetly during the period September–December. The averaged CO_2 seasonal amplitude was up to 15 ppmv in Gosan.

Figure 6.7 also shows seasonal cycles in Yanbian and Seoul, with maxima occurring in April and December respectively, with a minimum occurring in August at both sites. The averaged CO_2 seasonal amplitude was up to 18 ppmv in Seoul and 20 ppmv in Yanbian.

The seasonal variation of Seoul and Yanbian reflects the periodicity of terrestrial vegetation growth in the middle of NH.

6.4 Potential Source Region and Relative Emission Strength

The following figure shows the potential pollution source region and its relative emission strength derived from the hybrid receptor model (Fig. 6.8). Deduced from our model, the plain regions in northern China could be a potent source region for CO_2 in Gosan. The maximum relative emission strength is about 16 ppmv above background concentration.

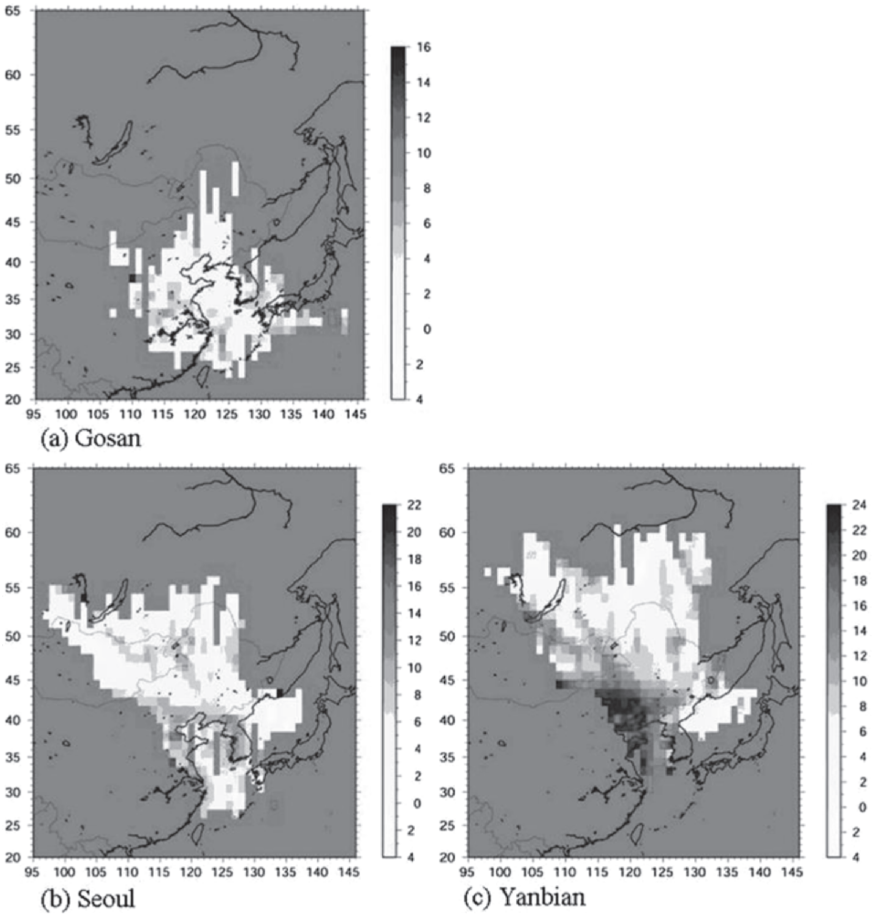

Fig. 6.8 CO_2 average concentration above background (ppmv), red color means high relative emission strength, yellow color means low relative emission strength. Units show concentration above background

During the sampling period, Seoul was mostly affected by sources located in eastern China and inside the Korean peninsula. The maximum relative emission strength is about 22 ppmv above background concentration. Yanbian was mostly affected by sources located in eastern China and inside the Korean peninsula. The maximum relative emission strength is about 22 ppmv above background concentration. Yanbian was mostly affected by sources located in eastern China. The maximum relative emission strength is about 24 ppmv above background concentration. The relative strength in Yanbian appears to be stronger than in Seoul.

The relative strength in Gosan is appears to be weakest, and the values in Yanbian to be the strongest.

In this study, the region around Shanghai, one of the most densely industrialized regions in Northeast Asia shows relatively low relative emission strength because the prevailing winds over the site at Gosan are rarely flowing from there. Though the hybrid receptor model has proven to be effective in identifying pollution sources, it has some limitations. Some of the modeling results show an unexpectedly large source pollution coming from the oceans. A possible explanation is that the trajectories with high concentration pass over the oceans. Further refinement of the model will help to explain or solve this problem.

6.5 Conclusions

We report the results of continuous atmospheric CO_2 measurements in the period from April 2004 to March 2005 at Gosan station, and in 2005 at Yanbian and Seoul stations using high-quality sampling data, by using the high-frequency high-precision NDIR CO_2 analysis system. Although measurement data through frequent flask sampling can show seasonal variations, it is difficult to know detailed variations such as diurnal variation and CO_2 concentrations affected by air mass transport on an hourly timescale. Pollution episode data and background data were separated using the AGAGE statistical method.

Atmospheric CO_2 background concentrations measured at Gosan, Seoul and Yanbian in recent years show typical diurnal and seasonal variations, higher values in the nighttime and lower values in the daytime, higher values in winter and lower values in summer. This is due to the terrestrial biosphere in NH and to photosynthesis by plants, and to the dilution of the boundary layer, etc.

Potential source regions in Northeast Asia were observed for anthropogenic atmospheric CO_2 by applying trajectory statistics. The results show the possibility of CO_2 potential sources in the plain regions in northern China contributing to Gosan, while contributions from northern and eastern China seem to be detected at Seoul and Yanbian. Seoul and Yanbian stations are important in creating a full potential source region map of the Northeast Asia region, as observations from Gosan are limited by the air masses transported to Gosan.

Further refinement of the modeling method will improve the accuracy of the potential source regions and relative emission strengths. With these improvements, and as atmospheric CO_2 data is accumulated over longer periods, we will be able to discuss long-term trends in the potential source regions and relative emission strengths in the Northeast Asia region.

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