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Temporal and spatial variations of the atmospheric CO_2 concentration in China

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[1] In order to understand the variations of atmospheric greenhouse gases over the Chinese mainland, an air sampling network was established in March 2003 that included one GAW global station, three GAW regional stations and three cooperative stations. Flask air samples were taken every week at the stations for 3 years. The secular increase and the seasonal variation in the CO_2 concentration are clearly observable at all the stations, reflecting global as well as the regional terrestrial biospheric and human activities, as well as the local meteorological conditions. The average CO_2 concentration depends on the station, with the lowest value observed at Mt. Waliguan and relatively high values observed at Fukang and Lin-an stations, located in the westernmost and eastern parts of China, respectively. The observed CO_2 concentration variations are also discussed within the context of forward transport model simulations using CO_2 fluxes derived by time-dependent inversion. **Citation:** Zhang, D., et al. (2008), Temporal and spatial variations of the atmospheric CO_2 concentration in China, *Geophys. Res. Lett.*, *35*, L03801, doi:10.1029/2007GL032531.

1. Introduction

[2] Precise measurements of the atmospheric CO_2 concentration over a wide geographical area are indispensable for quantifying its sources and sinks on the earth's surface [*Tans et al.*, 1989, 1990; *Fan et al.*, 1998; *Bousquet et al.*, 2000; *Patra et al.*, 2005a, 2005b]. The atmospheric CO_2 concentration has been monitored systematically at many places around the world since the 1950's [*Keeling et al.*, 1976a, 1976b; *Conway et al.*, 1994]. However, the current global CO_2 network is heavily biased toward ocean area. For a better understanding of the global carbon cycle, it is necessary to increase the number of CO_2 monitoring sites in the interior of continents. Indeed, additional inland stations have recently been established in Europe (AEROCARB, http://www.ipsl.jussieu.fr/~mrsce/index.html) and North America (The North American Carbon Program Plan, http://www.nacarbon.org/nacp/index.html). In China, flask air sampling was first started by the Chinese Academy of Meteorological Sciences (CAMS) at Mt. Waliguan (one of the 24 World Meteorological Organization/Global Atmosphere Watch (WMO/GAW) global baseline stations) in May 1990, in cooperation with the National Oceanic and Atmospheric Administration/Climate Monitoring and Diagnostics Laboratory (NOAA/CMDL), and has continued to the present [*Tang et al.*, 1995; *Zhou et al.*, 2003]. To reveal temporal and spatial variations of atmospheric CO_2 in the Chinese mainland in more detail, the CAMS, the Institute of Atmospheric Physics, Chinese Academy of Sciences and the Center for Atmospheric and Oceanic Studies, Tohoku University, Japan established a network of 7 monitoring sites that includes the Mt. Waliguan station, 3 GAW regional stations and 3 cooperative stations, and started CO_2 flask sampling in March 2003.

[3] In this paper, we describe our CO_2 concentration measurements in China and discuss the results obtained during the first 3 years of the program, especially in terms of seasonal cycle and annual average value of the CO_2 concentration. The observed CO_2 variations are compared with those simulated using an atmospheric general circulation model and 22-region time-dependent inversion CO_2 fluxes.

2. Experimental Procedures

[4] Our sampling network consists of 7 sites, as shown in Figure 1. Mt. Waliguan station (36.3°N, 100.9°E, 3810 m) is situated on the northeastern edge of the Tibetan Plateau. The surrounding area is arid/semi-arid grassland and desert steppe, and there are no industrial or pollution source areas in a radius of 50 km. Therefore, the Waliguan station is a suitable site for monitoring the background CO_2 concentration in the Eurasian Continent. Three stations in Longfengshan, Shangdianzi and Lin-an are approved as WMO/GAW regional stations. The Longfengshan station (127.6°E, 44.7°N, 310 m) is located in the northeastern part of China, with temperate forests and grasslands dominating its surrounding area. The Shangdianzi station (117.1°E, 40.4°N, 293.9 m) is 150 km northeast of Beijing, and orchard and farmland surround the station. The Lin-an station (119.1°E, 30.3°N, 138 m) is situated near the Yangtze River delta, with the surrounding area characterized as subtropical forests and farmland. Two of the 3 cooperative stations are the meteorological observatory at the summit of

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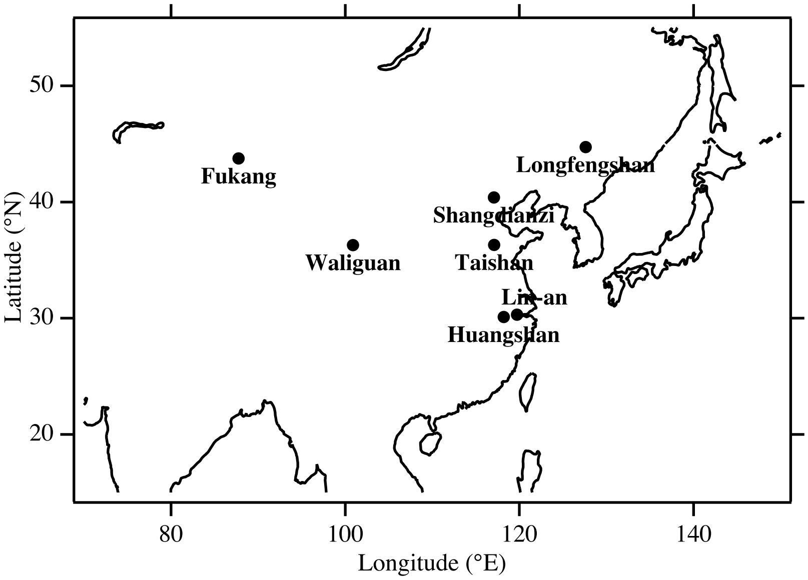

Figure 1. Map showing 7 sampling sites in China.

Mt. Taishan (117.1°E, 36.3°N, 1534 m) and Mt. Huangshan (118.2°E, 30.1°N, 1841 m). The remaining station is the observatory of the Chinese Academy of Sciences in Fukang (87.9°E, 44.3°N, 630 m) in the westernmost part of China, and the typical land surface around the station is oasis and desert.

[5] To collect air samples once per week at the stations, we have used two sets of instruments, one is 2.5 L Pyrex glass flasks and a sampling unit [*Thoning et al.*, 1995] from the NOAA/Earth System Research Laboratory (ESRL) and the other is 1.0 L stainless steel flasks and a sampling unit from Tohoku University. Both sampling units are equipped with a diaphragm pump to pressurize ambient air into the flask. The NOAA instrument was used at Waliguan, Longfengshan and Lin-an, with sample inlet located 5 m above the ground; each flask was pressurized to about 1.3×10^3 hPa. The Tohoku University instrument was used at Shangdianzi, Taishan, Huangshan and Fukang, and air samples collected at 10 m above the ground were pressurized into flasks at about 2×10^3 hPa. The glass and stainless steel flasks were rinsed by flowing ambient atmosphere through them at a rate of 5–9 L min^{-1} for about 15 min and at a rate of 10 L min^{-1} for about 3 min, respectively, to ensure a complete air exchange in the flasks. The collection of air samples was usually made early in the afternoon to obtain fully mixed air. The air sampling at each surface site was conducted as much as possible when winds were from areas not influenced by local human infrastructures and heavy vegetation.

[6] The collected air samples from the stations were sent to the Key Laboratory for Atmospheric Chemistry, CAMS in Beijing, and their CO_2 concentrations were determined with a precision of 0.1 ppm against our working standard gases using a non-dispersive infra-red gas analyzer (LI-COR 7000) within 1 month of sampling. To minimize the influence of water vapor on the measured CO_2 concentration value, each air sample was dried by passing it through a water vapor trap cooled to −50°C before introducing it into the analyzer. The working standard gases were CO_2-air mixtures stored in 30 L aluminum high-pressure cylinders. Their CO_2 concentrations were calibrated against the WMO-95 scale using 5 secondary standard gases with CO_2 concentrations of 348.09, 366.60, 379.09, 400.89 and 419.98 ppm which were purchased from the NOAA/ESRL in 2004. Each air sample was analyzed twice and their average was regarded as the sample CO_2 concentration.

[7] To extract the seasonal cycle, average value and increase rate of the CO_2 concentration, a digital filtering technique including Fourier harmonics, Reinsch-type cubic spline and Butterworth filter was applied to the CO_2 time series. Details of the technique have been described by *Nakazawa et al.* [1997]. In this study, the average seasonal CO_2 cycle was approximated by fundamental annual cycle and its first harmonics, and the cut-off period of the 26th-order Butterworth filter was chosen to be 24 months to determine the secular CO_2 trend. After the best-fit curve to the observed CO_2 values was obtained, the data lying more than 3 standard deviations from the curve were rejected from the dataset as outliers. The digital filtering was then conducted on the remaining data. These steps were repeated once or twice until no further outliers were found. The procedure rejected 2 to 8 data points as outliers for each site, corresponding to 1 to 6 % of the total data. The standard deviation of selected data from the best-fit curve (1σ) for each site is shown in Table 1.

3. Results and Discussion

[8] The CO_2 concentrations observed at the 7 sites are shown in Figure 2. It is obvious from Figure 2 that the CO_2 concentration increases with time at all sites, accompanying by clear seasonal cycle. The average CO_2 increase rates derived from the best-fit curves for the respective sites range between 3.6 and 1.7 ppm yr^{-1}, as summarized in Table 1. The different CO_2 increase rates observed in this study can be attributed to different influences of regional CO_2 sources and sinks, air transport and local meteorology at individual stations. In this regard, it is worthy of note that the emission of fossil fuel CO_2 in China has been increasing significantly in recent years [*Marland et al.*, 2005] due to rapid economic

Table 1. Average Increase Rates, Average Values and Seasonal Cycles of the CO_2 Concentration, and Standard Deviations of the Observed Data From the Best-Fit Curve at 7 Sites in China

Site	Observation Periods	Increase Rate, ppm yr^{-1}	Average Conc., ppm	Seasonal Maximum, ppm	Appearance of Seasonal Maximum, day	Seasonal Minimum, ppm	Appearance of Seasonal Minimum, day	Peak-to-Peak Amplitude, ppm	Standard Deviation, ppm
Fukang	Apr. 2003 – Apr. 2006	2.4	385.3	12.8	18	−12.0	222	24.8	3.4
Huangshan	Mar. 2003 – Mar. 2006	2.3	380.3	6.2	57	−7.1	236	13.3	3.2
Lin-an	Jun. 2003 – Jun. 2006	3.0	390.3	6.0	22	−5.5	235	11.5	8.1
Longfengshan	Jun. 2003 – Jun. 2006	3.6	383.8	8.0	25	−11.3	207	19.3	4.1
Shangdianzi	Jul. 2003 – May 2006	2.7	383.8	6.1	62	−9.9	214	16.0	6.4
Taishan	Apr. 2003 – Apr. 2006	1.7	382.0	8.0	42	−10.3	226	18.3	5.3
Waliguan	Jun. 2003 – Apr. 2006	1.9	378.4	5.1	86	−5.8	227	10.9	1.7

growth, resulting in the high CO_2 increase rates observed at our surface sites especially in eastern China.

[9] Characteristics of the average seasonal CO_2 cycle at individual stations are given in Table 1. The CO_2 concentration usually reaches a maximum in February and a minimum in August. However, the timing of the occurrences of seasonal maximum and minimum varies significantly from station to station, ranging from a maximum in January at Fukang to a maximum at the end of March or in early April at Waliguan, while the seasonal minimum at Longfengshan is found in July and at Shangdianzi at the end of July or in early August. These differences are produced not only by regionally different terrestrial biospheric and human activities but also by local meteorological conditions. The high CO_2 concentrations observed at Fukang in the wintertime is likely due to a local strong surface inversion that traps CO_2 emitted by human and terrestrial biospheric activities [*Li*, 1991]. A long-range air transport is important for the appearance time of the maximum CO_2 concentration at Waliguan, since its timing agrees well with those observed at many background monitoring sites at similar latitudes [e.g., *Nakazawa et al.*, 1993]. As seen in Figure 2, no summertime low CO_2 concentrations were observed at Lin-an in 2003. We speculate that this resulted from high CO_2 emitted by local electric power stations operating at maximum capacity during the hot/dry summer of 2003 to match the power shortage caused by the suspending of originally dominant hydro-power stations in Zhejiang Province where Lin-an station is located. It is interesting to note that the same region was struck by less intense heat waves in the summer of 2005, resulting in higher CO_2 concentration than that observed during the summer of 2004.

[10] The largest seasonal CO_2 cycle with a peak-to-peak amplitude of 24.8 ppm is found at Fukang, mainly due to the wintertime CO_2 accumulation in the surface inversion layer. On the other hand, Waliguan shows the smallest amplitude of 10.9 ppm, since it is a high mountain site. Lin-an also shows a small seasonal amplitude of 11.5 ppm, mainly due to the high summertime CO_2 concentrations especially in 2003 and 2005. The small seasonal cycle observed at Lin-an is also due to the fact that the station is located in the southern part of our network where seasonal variation in the biospheric flux is smaller. The seasonal amplitudes at the remaining stations range between 13.3 and 19.3 ppm, which are in agreement with those at other continental sites at similar latitudes. For example, the amplitude of 19.3 ppm at Longfengshan (44.7°N) is similar to those at Kasprowy Wierch, Poland (19.1°E, 49.2°N, 1987 m) [*Necki et al.*, 2003] and Fraserdale, Canada (81.6°W, 49.8°N) [*Higuchi et al.*, 2003]. However, the seasonal CO_2 cycles at Longfengshan and Taishan (36.2°N, 1534 m) are larger than those observed at similar latitudes in Japan located downwind of China; the respective amplitudes are larger by 1.6 and 4.9 ppm than those near Wakkanai (141.5°E, 45.5°N) (T. Nakazawa, Tohoku University, unpublished data, 2007) and at 0–2 km over Sendai (141.8°E, 37.7°N) [*Nakazawa et al.*, 1993] for this analysis period.

[11] The average CO_2 concentrations at the 7 sites are also listed in Table 1. Lin-an shows the highest value of 390.3 ppm, reflecting the fact that the summertime CO_2 concentrations were extremely high especially in 2003 and 2005 due to local contamination. The value at Fukang is also high, i.e. 385.3 ppm, mainly due to the wintertime accumulation of anthropogenic CO_2 near the ground surface. The average CO_2 concentrations at the mountain stations are lower than those at the surface stations, since the mountain sites are remote from areas with strong human activities. For example, Waliguan station shows the lowest CO_2 concentration of 378.4 ppm and the value at Huangshan is found to be 380.3 ppm. However, the average CO_2 value at Taishan is higher by 2–4 ppm than those at the other 2 mountain stations. In this connection, it may be noted that Mt. Taishan is situated in the North China Plain with a high population and intense industrialization. The NOAA/ESRL has also measured the CO_2 concentration at Waliguan (ftp://ftp.cmdl.noaa.gov/ccg/co2/flask/month/wlg_01D0_mm.co2). By comparing with their monthly mean values, we found that our results agree fairly well with theirs, except for summer months when our values are clearly lower by a few ppm. As a result, our average CO_2 concentration over the present observation period is lower by 0.7 ppm than theirs. The summer difference with the NOAA/ESRL could be due to sampling time; their samples were collected at around 8 a.m., while our sampling was made in the early afternoon. Continuous CO_2 measurements at Mt. Waliguan show a clear diurnal cycle in summer [*Zhou et al.*, 2003], probably due to diurnal effects of local biospheric activities and/or upslope/downslope winds.

[12] To understand how CO_2 sources and sinks contribute to the spatially different variations of the CO_2 concentration in China, we employ an atmospheric general circulation model (AGCM [e.g., *Takigawa et al.*, 2005]) and one-year monthly-mean CO_2 fluxes derived from a 22-region time-dependent inversion. This set of flux is estimated following *Gurney et al.* [2004], but using atmospheric CO_2 data [*GLOBALVIEW-CO_2*, 2005] from a network of 87 stations

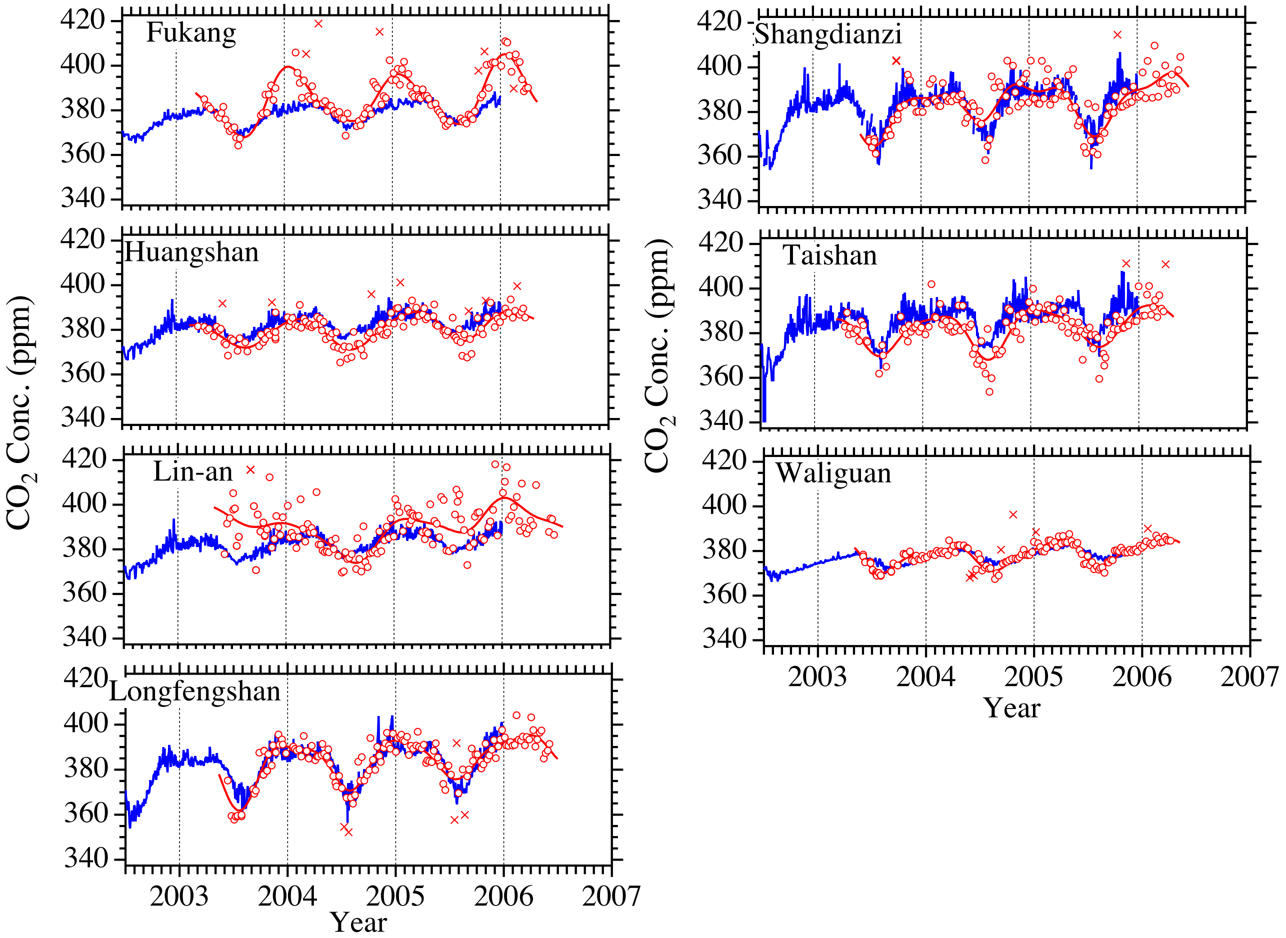

Figure 2. Temporal variations of the CO_2 concentration at 7 sites in China. Circles, solid lines, and crosses show the observed data, their best-fit curve, and outliers rejected in the curve fitting, respectively. The CO_2 concentration variations simulated using the atmospheric general circulation model and 22-region time-dependent inversion CO_2 fluxes are also shown by blue lines.

for the period 1999–2001 (see *Patra et al.* [2005a] for site locations). An average of inverse fluxes calculated by 12 atmospheric transport models is used in our AGCM simulation. The CO_2 concentrations thus calculated are also plotted in Figure 2 for each of the stations. In Figure 2, all the calculated concentration values are shifted so that the annual average of the calculated CO_2 concentration for 2005 at Waliguan agrees with the observed value. Because the model spin-up run was performed for a short period (5 years), an offset is required to match the observed concentration, and we selected Waliguan for the offset determination as CO_2 data from this site was utilized in the inverse calculation. It is seen that the CO_2 concentration variations calculated for Waliguan, Shangdianzi, Longfengshan and Huangshan are fairly consistent with the observed results, while the disagreement between the calculated and the observed CO_2 variations is clearly found at Fukang, Lin-an and Taishan. Especially at Fukang in the winter season, the calculated CO_2 concentrations are much lower than the observed values, suggesting that the model cannot reproduce the local effect of the anthropogenic CO_2 accumulation in the strong surface inversion layer in very cold season. The CO_2 concentrations calculated for Lin-an are also lower than the observed values, except for 2004; this is mainly due to neglecting the effect of additional CO_2 emissions that occurred in the summer of 2003 and 2005. On the other hand, the calculated CO_2 concentrations at Taishan are clearly higher than the observed values, especially in the warm season. A similar phenomenon is also seen at Huangshan, although the concentration differences are smaller. This discrepancy may suggest that the summertime upward air transport in the model is weaker than what actually takes place in the real atmosphere. Note here that the inverse model flux regions are too coarse to capture regional emission distributions, no interannual variability in fluxes is considered and in 22-region inverse model, China, Mongolia and two Koreas form one region.

4. Conclusions

[13] By systematically measuring the CO_2 concentration at 7 sites in China for almost 3 years from March 2003, we have presented its temporal and spatial characteristics over the Chinese mainland. The observed CO_2 variations were

reproduced reasonably well using the AGCM with 22-region time-dependent inversion CO_2 fluxes, although significant differences between the calculated and the observed CO_2 concentrations were found at stations with strong local effects on the CO_2 concentration. The atmospheric CO_2 concentration data obtained in this study would be useful for constraining CO_2 sources and sinks in China at higher spatial resolutions. Given the fact that China is emerging as a significant contributor to future atmospheric CO_2 levels, it is important to expand the observation network, bind it interactively closer together and maintain it for a long time, which will benefit future/high-resolution inverse modeling efforts.

[14] **Acknowledgments.** We express our sincere thanks to all the staffs who contributed to the collection of air samples at Waliguan, Shangdianzi, Longfengshan, Lin-an, Taishan, Huangshang and Fukang stations. This work was supported by the China Ministry of Science and Technology project (2001DIA10009), the research project 2-1 of the Research Institute for Humanity and Nature, Japan, and the Research Revolution 2002 (project 3) and the Grants-in-Aid for Creative Scientific Research (2005/17GS0203) of the Ministry of Education, Science, Sports and Culture, Japan.

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