

## EPISODIC ENHANCEMENTS OF CO<sub>2</sub> AND CO AT THE SUMMIT OF MT. FUJI (3776 M), JAPAN

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### ABSTRACT

The mixing ratios of atmospheric CO<sub>2</sub> were observed at the summit of Mt. Fuji by using a system for continuous measurements during September 2002-February 2003 and May 2003-May 2004. The observed CO<sub>2</sub> variations at Mt. Fuji showed a seasonal cycle of the background level with a maximum around April and a minimum around August. A lot of episodic events with a large enhancement of CO<sub>2</sub> were found, and the episodic enhancements of CO<sub>2</sub> at Mt. Fuji were well associated with increased CO peaks observed at the same time. The enhancement ratios of CO to CO<sub>2</sub> mixing ratios ( $\Delta\text{CO}/\Delta\text{CO}_2$ ) mainly showed lower values of less than 0.03 due to urban/industrial sources, while relatively higher  $\Delta\text{CO}/\Delta\text{CO}_2$  ratios up to 0.08 were also found for the episodic events due to the biomass burning emissions. Three-dimensional transport model simulations of CO suggested that the major contributions for the increased events at Mt. Fuji were from China (~50%) and the other major regions were Southeast Asia and South Asia (~10%).

### INTRODUCTION

A high-mountain observatory at the summit of Mt. Fuji (35°22'N, 138°44'E, 3776 m above sea level) in Japan is suitable for atmospheric chemistry study to observe trace gas variations in the free troposphere, because this mountain has a high and slim solo peak. Prevailing wind at the summit is usually westerly which is particularly strong in winter. This strong westerly winds indicate that Mt. Fuji is situated downwind of industrial emissions from the Asian continent to evaluate a direct influence of the Asian outflow on the free troposphere. We have made continuous measurements of CO<sub>2</sub> and CO at this weather station of JMA. Previously, continuous CO<sub>2</sub> measurements at this summit were made by Nakazawa *et al.* [1984] from July to October, 1981, but no other longer observations were conducted before. Thus, our observation was first to observe detailed CO<sub>2</sub> variations throughout a year at Mt. Fuji.

### MESURING SYSTEM

Outside air was taken from an intake located on the meteorological tower (about 10 meters above the summit) by a diaphragm pump and then dried using an electric dehumidifier and a chemical desiccant (Mg(ClO<sub>4</sub>)<sub>2</sub>). The CO<sub>2</sub> mixing ratios were measured using an NDIR (LI-COR, type LI-6252). The pressure-control device and mass-flow controller were used to maintain constant pressure and flow rate. The CO<sub>2</sub> measuring system was calibrated every 3 hours by using 3 standard gases of CO<sub>2</sub> in air. They were prepared in 10 L high-pressure cylinders by a gas company (Nihon Sanso Co. Ltd., Japan and Taiyo Toyo Sanso Co. Ltd., Japan). The CO<sub>2</sub> mixing ratios in the cylinders were determined using the primary standard gases as used for MRI-87 scale reported in Inoue and Matsueda [2001]. The mixing ratios of atmospheric CO<sub>2</sub> were observed at the summit of Mt. Fuji by using this system during September 2002-February 2003 and May 2003-May 2004. The simultaneous CO measurements were conducted by using a gas-filter correlation method (TECO, model 48C) from September 2000 to July 2004.

### CO<sub>2</sub> OBSERVED AT MT. FUJI

Time series of observed CO<sub>2</sub> mixing ratios at Mt. Fuji were shown in Fig. 1. The observed CO<sub>2</sub> variations at Mt. Fuji showed a seasonal cycle of the background level with a maximum around April and a minimum around August. A similar pattern of CO<sub>2</sub> seasonal cycle was commonly observed in the JMA and NOAA/CMDL ground-based stations in the northern mid-latitudes [Conway *et al.*, 1994, WDCGG, 2004], although the monthly means of CO<sub>2</sub> at Mt. Fuji were slightly higher in winter (~3 ppm) and lower in summer (~2 ppm) compared with CO<sub>2</sub> observed at

Minamitorishima (24°18'N, 153°58'E) in western North Pacific. A lot of episodic events with a large enhancement of CO<sub>2</sub> were found at Mt. Fuji as shown in Fig. 1. The episodic events occurred at an interval of less than ten days and often lasted a half-day to several days. No significant diurnal cycle was found in the CO<sub>2</sub> at Mt. Fuji, indicating that the episodic enhancement events were not caused by the local pollution due to urban/industrial emissions in Japan, but they were relatively large-scale phenomena caused by the continental outflow. The episodic enhancements of CO<sub>2</sub> at Mt. Fuji were well associated with increased CO peaks observed at the same time. The enhancement ratios of CO to CO<sub>2</sub> mixing ratios ( $\Delta\text{CO}/\Delta\text{CO}_2$ ) mainly showed lower values of less than 0.03 due to urban/industrial sources, while relatively higher  $\Delta\text{CO}/\Delta\text{CO}_2$  ratios up to 0.08 were also found for the episodic events due to the biomass burning emissions.

### NUMERICAL SIMULATIONS FOR EPISODIC POLLUTED EVENTS

In order to quantitatively evaluate these episodic polluted events at Mt. Fuji, three-dimensional transport model simulations of CO variations were performed using the EDGAR/GEIA emission database [Oliver *et al.*, 1999]. Detailed information of the basic procedures for the simulations was reported by Taguchi *et al.* [2002]. The model almost reproduced the observed pollution events with the enhanced CO peaks. To evaluate the individual contributions on the variations at Mt. Fuji, other simulations were performed separately for 13 different emission regions. During the episodic events, the CO from emissions in China increased about 4 times as high as the non-events, indicating that the CO emitted from China accounted for about a half of the CO enhancement peaks in the model. On the other hand, the CO from emissions in Southeast Asia and South Asia during the episodic events was higher by 2-3 times than that during the non-events, and they accounted for 6-11 % of the total enhancement of CO. The time-altitude cross sections over Mt. Fuji in the model showed several different transport pathways for the outflow from the Asian emission regions. The CO-enriched plumes from the China occurred in the both boundary layer and free troposphere, while the plumes from Southeast Asia and South Asia regions were found only in the free troposphere. It was also found that no CO-enriched plumes in the free troposphere were found in the cross sections for North America and Europe throughout a year, while the influences from emissions in these regions are thought to be effective to the background CO seasonal cycle at Mt. Fuji.

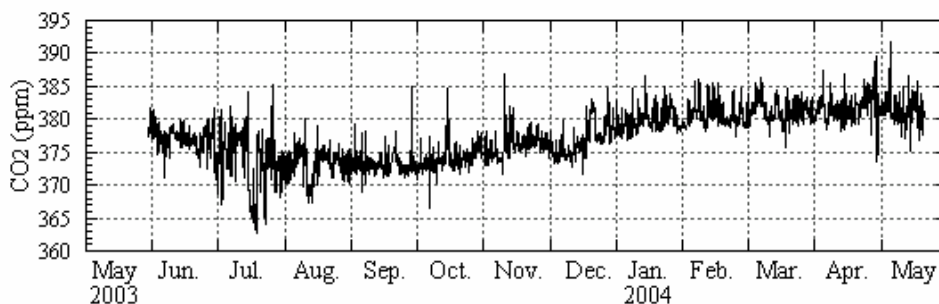


Fig. 1. CO<sub>2</sub> mixing ratios observed from May 2003 to May 2004 at the summit of Mt. Fuji, Japan.

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