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Observations of carbon monoxide mixing ratios at a mountain site in central Taiwan during the Asian biomass burning season

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ABSTRACT

Carbon monoxide (CO) mixing ratios were observed from 30 January to 7 April 2008 at Mt. Lulin (23.51°N, 120.92°E, 2862 m asl) in central Taiwan to investigate characteristics of CO during biomass burning periods. During the sampling campaign, the average mixing ratio of CO was 234 ± 63 ppb with higher levels observed in March. The elevated CO in March can, on the basis of backward trajectories and satellite fire spots analyses, possibly be attributed to biomass burning activities in the Asian continent. Significant diurnal variations of CO mixing ratios were observed at the remote site. The higher CO levels in the afternoon were influenced by the transport of boundary layer pollution to the site during daytime upslope flow. Backward trajectory analysis showed that air masses mainly originated from India (ID), the Indochina Peninsula (IP) and South Coastal China (SC), which together accounted for 85% of the total trajectories. Higher mixing ratios of CO were found in the ID, IP, and SC categories, indicating significant impacts of anthropogenic emissions on the Pacific region. Furthermore, the air parcels were divided into two categories, those that passed over the fire regions and those that did not. The result showed that the average difference of CO levels between the two categories was approximately 79 ppb, suggesting that Asian biomass burning plays an important role in CO levels at this remote site during the springtime.

1. Introduction

Carbon monoxide (CO) plays an important role in the oxidation chemistry of the troposphere. CO oxidation leads to the formation of hydrogen peroxy radicals (HO_2), which can react to produce O_3 in the NO_x rich environment. On the contrary, CO oxidation, caused by its reacting with OH, leads to O_3 destruction through the HO_x catalytic cycle in the low level NO_x atmosphere (Kanakidou and Crutzen, 1999). Changes in OH mixing ratios due to changes in CO mixing ratios can perturb the mixing ratios of greenhouse gas as methane (CH_4) (Thompson and Cicerone, 1986). Thus CO, not in itself a "greenhouse" gas, can indirectly contribute to global warming because of its important role in tropospheric OH chemistry. Natural and anthropogenic sources contribute almost equally to the observed mixing ratios of CO, suggesting that combustion of fossil fuels, biomass burning, and oxidation of methane are responsible for approximately 85% of the total CO emissions (Oliver et al., 1999).

Biomass burning activity is recognized as an important source of CO and other trace gases and recently became a hot topic in global atmospheric chemistry. The activities are usually seen in tropical regions, such as Southeast (SE) Asia, South America, and Africa, and even Northern Australia is well known to be an extensive biomass burning area (Thompson et al., 2001; Boian and Kirchhoff, 2004; Meyer et al., 2008). Biomass burning in SE Asia is mostly caused by deforestation and agricultural activities. However, India, Thailand, Myanmar and Vietnam are the most active regions of biomass burning in the SE Asian continent (Streets et al., 2003). The evidence of biomass burning resulting in enhanced CO in the downstream

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regions has been examined by observations and model simulations. Pochanart et al. (2003) suggested the long-range transport of air masses driven by biomass burning activities in continental SE Asia was the main factor of increased CO mixing ratio during the springtime in Thailand. Tang et al. (2003) reported that the biomass burning net influence of CO was as high as 50–100 ppb for the layer below 3 km over the Pacific region.

Mountain-top free tropospheric observation sites are sparsely located in the northern hemisphere. They are generally located far away from the direct influence of the polluted plume. Due to the high elevation, the pollutant levels are often determined by transport process on a regional scale (Weiss-Penzias et al., 2006). During January 30 to April 7, we conducted CO measurements on the summit of Mt. Lulin, located in central Taiwan. This mountain-top location was selected for this study because its high altitude allows the collection of long-range transported pollutants. At the same time, the weather parameters (wind fields, temperature, relative humidity, and rainfall) were also continuously observed by the Taiwan Environmental Protection Administration (EPA). In this paper, we report the characteristics of CO during the biomass burning season. The comparisons of this study with other measurements in the world are also discussed. The influence of the various air parcel transport pathways on CO mixing ratios is also investigated based on backward trajectory analyses. Finally, the net influence of SE Asian biomass burning on CO mixing ratio is assessed in this study.

2. Methods

2.1. CO measurement

The CO measurements have been made at the summit of Mt. Lulin (23.51°N, 120.92°E), 2862 m above sea level (asl). Mt. Lulin is part of the Central Range and is located in the Yushan National Park as shown in Fig. 1. The nearest major city, Taichung, is about 90 km to the northwest and has a population of 1,000,000. There is no major city within a 50-km radius from the sampling site, only small villages spread sparsely over the range. The nearest residences to the observation site are those in Tungpu village located downhill at a distance of 15 km at the elevation of 1200 m. The population of the small village is about 1300. Because of the high elevation, Mt. Lulin is almost completely isolated from local pollution, and therefore it has been postulated to be a good site to observe the long-range transported pollutants such as the biomass burning plumes from the SE Asian continent. During the measurements, the instrument was housed in a container at the Lulin station. Ambient air was drawn through a Dekoron sampling tube (1/4 in. O.D. Synflex, USA) with the inlet placed 10 m above the ground (4 m above the rooftop of the container). Inside the laboratory, the sampling tube was connected to a glass manifold with a bypass pump to reduce the residence time of sampled air.

CO mixing ratio was measured by using a commercial vacuum-UV resonance fluorescence (RF) CO instrument (AEROLASER GmbH, model AL-5002) with high precision and time resolution. This technique was first developed by Volz and Kley (1985) in the laboratory and recently applied to low CO mixing ratio measurements, especially in aircraft observations (Takegawa et al., 2001). It consists of a resonance lamp excited by an RF discharge, an optical filter for selecting the appropriate wavelength interval around 150 nm, which images the lamp into the RF chamber, where the fluorescence is viewed by means of a photomultiplier. The resonance lamp was operated with a mixture of 0.25% CO_2 in high pure Ar (Scott, 6 N) at a flow rate of 35 ml min^{-1}. The optical filter was gently flushed with 25 ml min^{-1} dry nitrogen (Praxair, 6 N) to avoid absorption of oxygen and other impurities. For N_2 an additional purifier (Entegris, 500KF) was used to remove water and hydrocarbons. As the method has a slight sensitivity to water, the sample gas had to be dried before it entered the fluorimeter. The dry gas was circulated through an integrated Nafion dryer containing a small pump and desiccant. Before the sampling campaign, we checked the linearity of the instrument with the internal zero gas and five NIST traceable CO standards (65, 152, 244, 291, and 487 ppb), purchased from Scott-Marrin Inc., California in the laboratory. The results showed that the correlation coefficient (r^2) of the linear regression was 0.999. During the measurement, daily instrument checks were performed by using a 487 ppb CO standard and zero internal gas. For determination of the zero signals, the CO standard passed through a zero trap with Sofnocat catalyst, which quantitatively removed the CO < 1 ppb. The detection limit of this instrument was 1.5 ppb for a 1-min average with precision of ±1% for a level of 487 ppb (1-min average), and the overall uncertainty was within 1.5% during the sampling periods.

2.2. Backward trajectory analysis

To identify the origins and the transport paths of air masses prior to their arrival at Mt. Lulin, the 5-day backward trajectories were calculated by using the HYbrid Single-Particle Lagrangian Integrated Trajectory (HYSPLIT) model developed by USA NOAA Air Resources Laboratory (Draxier and Hess, 1998). The meteorological input for the trajectory model was the GDAS (Global Data Assimilation System). The GDAS data were processed by the National Centers for Environmental Predictions (NCEP) with a 6-h time resolution, about 190 km horizontal resolution, and 23 vertical levels. In this research, backward trajectory was calculated at a frequency of 6 synoptic times per day, i.e., at 0000, 0400, 0800, 1200, 1600 and 2000 LT (local time), with an initial altitude level of 3000 m, and a time step of 6 h. Additional 4 trajectories of which starting locations were changed ±0.5° from the actual sampling site to reduce the uncertainty of the trajectory analysis.

2.3. Satellite images of fire spots

The fire spots from biomass burning were extracted by the Along Track Scanning Radiometer (ATSR) satellite data developed by the European Space Agency (ESA). The ATSR instrument that provided the data set had four visible and near infrared channels centered at 0.55, 0.65, 0.86, and 1.6 μm, together with mid-infrared and thermal-infrared channels centered at 3.7, 10.8, and 12 μm. The ATSR