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Characteristics of atmospheric carbon monoxide at a high-mountain background station in East Asia

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HIGHLIGHTS

- 5-year real-time measurements of surface CO at a mountain-top station in East Asia.
- Generally coherent results were found between in-situ CO data and NOAA flask data.
- Distinct seasonal and diurnal cycles were observed and discussed in depth.

Keywords:
CO
Lulin atmospheric background station (LABS)
Biomass burning
Background values

ABSTRACT

Atmospheric CO were monitored at the Lulin Atmospheric Background Station (LABS) with an elevation of 2862 m AMSL from April 2006 to April 2011 by the in-situ non-dispersive infrared (NDIR) spectrometer and weekly flask sample collections via collaboration with NOAA/ESRL/GMD. In general very coherent results were observed between the two datasets, despite a slight difference between the two. A distinct seasonal pattern of CO was noticed at the LABS with a springtime maximum and a summertime minimum, which was predominately shaped by the long-range transport of biomass burning air masses from Southeast Asia and oceanic influences from the Pacific, respectively. Diurnal cycles were also observed at the LABS, with a maximum in late afternoon and a minimum in early morning. The daytime CO maximum was most likely caused by the up-slope transport of lower elevation air. After filtering out the possibly polluted data points from the entire dataset with a mathematic procedure, the mean background CO level at the LABS was assessed as 129.3 ± 46.6 ppb, compared to 149.0 ± 72.2 ppb prior to the filtering.

The cluster analysis of the backward trajectories revealed six possible source regions, which shows that air masses originating from the Westerly Wind Zone were dominated in spring and winter resulting in higher CO concentrations. As a contrast, the oceanic influences from the Pacific were found mostly in summer, contributing a lower seasonal CO concentration throughout a year.

1. Introduction

Carbon monoxide (CO) is an important trace gas which alters the oxidizing capacity of the atmosphere indirectly by reacting with hydroxyl radical (OH) in the troposphere (Logan et al., 1981; Thompson, 1992). It also acts as an intermediate in the oxidation pathways such as in the destruction and combustion of reduced compounds to ultimately produce carbon dioxide (CO_2) (Brenninkmeijer and Novelli, 2003). Under the condition of low levels of non-methane hydrocarbons (NMHCs), CO consumes about 75% of the available OH while methane (CH_4) consumes most of the rest (Thompson, 1992). Though CO is not a greenhouse gas, an increase of CO may depress the OH concentration in the atmosphere,

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and consequently, lead to less removal of CH_4 (Thompson and Cicerone, 1986). Therefore CO exerts an indirect effect on global warming through its oxidative reactions.

Both natural and anthropogenic sources, including fossil fuel combustion, biomass burning, and oxidation of CH_4 and NMHCs, contribute to the CO level in the atmosphere (Logan et al., 1981; Holloway et al., 2000). For instance, in 1998, 2002, and 2003, significant CO increases in the northern hemisphere were found to be associated with anomalously large forest fires (Novelli et al., 2003; Yurganov et al., 2004, 2005; Duncan et al., 2007). These burnings produce large amounts of CO and other ozone precursors, and ozone can be formed in the downwind area of the source region (Fishman and Seiler, 1983; Cros et al., 1988; Novelli et al., 1998; Chalbot et al., 2013). Duncan et al. (2007) have summarized the budgets for CO sources calculated by individual efforts, showing a range of 20–25% of global CO that is contributed by biomass burning activities. In tropical Asia, extensive fire spots detected by satellite over the Southeast Asian subcontinent have been reported (Christopher et al., 1998; Ito and Penner, 2004; Giglio et al., 2006; Wang et al., 2007). Cooperative campaigns, e.g., Transport and Chemical Evolution over the Pacific (TRACE-P) (Jacob et al., 2003; Tang et al., 2003), Asian Pacific Regional Aerosol Characterization Experiment (ACE-Asia) (Huebert et al., 2003), Biomass-burning Aerosols in South-East Asia: Smoke Impact Assessment (BASE-Asia) (Tsay et al., 2013), and Seven SouthEast Asian Studies (7-SEAS) (Lin et al., 2013; Reid et al., 2013), were conducted to study the atmospheric chemistry and radiation properties influenced by the biomass burning over this region. In addition to biomass burning, rapid urbanization and industrialization in East Asia in recent years have raised growing concerns over the impacts of Asian continental outflows on the air quality of downwind areas (Akimoto et al., 1996; Jacob et al., 1999; Bey et al., 2001; Heald et al., 2003; Liang et al., 2004; Jaffe et al., 2005; Hatakeyama et al., 2011; Lin et al., 2012; Tsai et al., 2012). With a lifetime of 1–3 months, CO can be transported inter-continentally (Jennings et al., 1996).

The Lulin Atmospheric Background Station (LABS), located at a mountain top in central Taiwan, is an ideal site to monitor the changes in baseline concentrations of various key pollutants (Wai et al., 2008; Sheu et al., 2010; Lee et al., 2011; Ou-Yang et al., 2012) released from the Asian continent into the Pacific Basin. The availability of the observations at the LABS was timely considering the rapid social and economic developments in this region in the past decades. With multiple origins of air masses arriving at the LABS site throughout a year, the characteristics associated with various source regions can be distinct and hence accentuate the uniqueness of this site. In this study, we present five-year continuous measurements of CO conducted at the LABS from the perspectives of variations in CO concentrations and the mechanisms that drive the variations.

2. Experimental

2.1. Site description

Fig. 1 shows the geographical location of the LABS (23.47°N, 120.87°E; 2862 m AMSL; http://lulin.tw/) associated with the averaged trajectories based on a cluster analysis (see further discussion in Section 3.4). The LABS is a two-story building situated at the peak of Mt. Front Lulin in Yu-Shan National Park in central Taiwan. There are no known strong emissions in the proximity of the station. The LABS is frequently within the free troposphere and is therefore an ideal site for conducting measurements of long-range transport of air pollutants, complementing the global network of the Global Atmospheric Watch (GAW) in the East Asia region where no other high-elevation baseline station is available. Further detailed descriptions of the LABS can be found in the literature (Sheu et al., 2010).

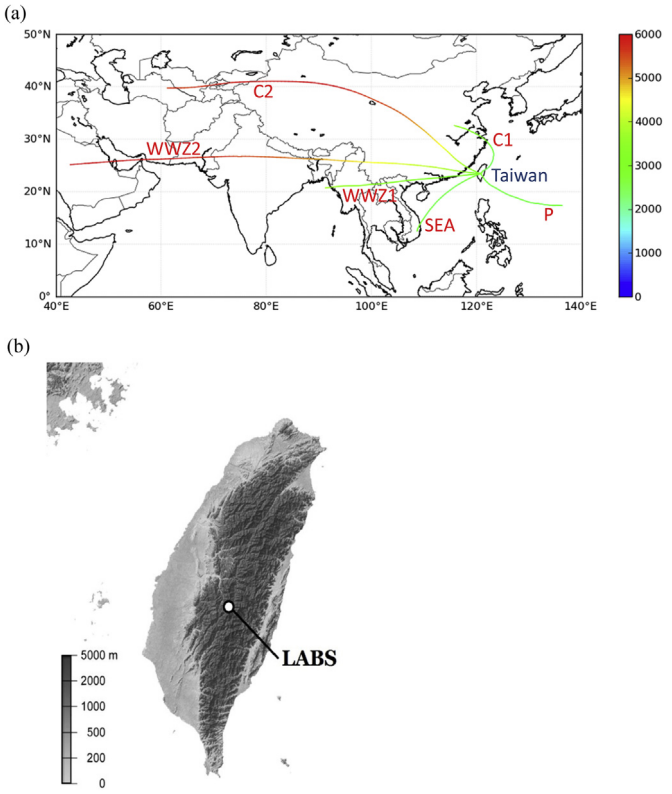

Fig. 1. (a) Geographic location of the LABS associated with classified mean backward trajectories and (b) terrain map.

2.2. Measurements

All of the gas analyzers, including the CO and other gas instruments, are located on the second floor of the building, with the air intake line extruding through the roof with the inlet point approximately 10 m above ground. An oil-free fan provided a gas flow rate of approximately 2.0 m^3 min^{-1} to the gas analyzers. The CO mixing ratio was obtained by an NDIR (APMA-360, Horiba, Japan) at the LABS from April 2006 to April 2011. The LABS data can be assessed at http://lulin.tw/index_en.htm. The instantaneous data points were taken at a frequency of 6 s and further calculated into hourly averages. The detection limit of the NDIR is ~20 ppb (1σ) (Zellweger et al., 2009), and the overall uncertainty is calculated to be ±14.4 ppb based on the mean standard deviation of the 6 s data within each hour in this study. A pair of 2L flask samples was also collected weekly and analyzed by NOAA/ESRL/GMD using gas chromatography/reduction gas detection (GC-RGD) or vacuum ultraviolet/resonance fluorescence (VUV-RF) method since August 2006. The overall uncertainty of the CO measurements by NOAA/ESRL/GMD during this period is estimated to be ±1.4 ppb. Further detailed descriptions about the sampling and analytical method can be found on the NOAA/ESRL/GMD's webpage: http://www.esrl.noaa.gov/gmd/outreach/behind_the_scenes/measurementlab.html.

2.3. Calibration

To calibrate NDIR a dynamic dilutor (Model 4010, Sabio, USA) with a cylinder standard filled with ~10 ppmv CO (Scott–Marine, USA) was used on site as the span source to make monthly multi-point calibration checks in the range of 0–800 ppb. The accuracy of the calibration points was checked by GC-RGD whose calibration is discussed later in the text. The Scott–Marine CO standard was renewed every 3 years. A supposed CO-free air stream made by a