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Aerosol transport from Chiang Mai, Thailand to Mt. Lulin, Taiwan – Implication of aerosol aging during long-range transport

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HIGHLIGHTS

- $PM_{2.5}$ levels after being transported 2400 km are lowered by a factor of 3.5
- The modification of an aerosol component during long-range transport was assessed.
- Most water-soluble components are enhanced during long-range transport.
- Biomass burning aerosol is more water-soluble in the downwind than source region.
- Anhydrosugars were found degraded during long-range transport.

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ABSTRACT

The transport of biomass burning (BB) aerosol from Indochina may cause a potential effect on climate change in Southeast Asia, East Asia, and the Western Pacific. Up to now, the understanding of BB aerosol composition modification during long-range transport (LRT) is still very limited due to the lack of observational data. In this study, atmospheric aerosols were collected at the Suthep/Doi Ang Khang (DAK) mountain sites in Chiang Mai, Thailand and the Lulin Atmospheric Background Station (Mt. Lulin) in central Taiwan from March to April 2010 and from February to April 2013, respectively. During the study period, an upwind and downwind relationship between the Suthep/DAK and Lulin sites (2400 km apart) was validated by backward trajectories. Comprehensive aerosol properties were resolved for $PM_{2.5}$ water-soluble inorganic ions, carbonaceous content, water-soluble/insoluble organic carbon (WSOC/WIOC), dicarboxylic acids and their salts (DCAS), and anhydrosugars. A Modification Factor (MF) is proposed by employing non-sea-salt potassium ion (nss-K^+) or fractionalized elemental carbon evolved at 580 °C after pyrolized OC correction (EC1-OP) as a BB aerosol tracer to evaluate the mass fraction changes of aerosol components from source to receptor regions during LRT. The MF values of nss-SO_4^{2-}, NH_4^+, NO_3^-, OC1 (fractionalized organic carbon evolved from room temperature to 140 °C), OP (pyrolized OC fraction), DCAS, and WSOC were above unity, which indicated that these aerosol components were enhanced during LRT as compared with those in the near-source region. In contrast, the MF values of anhydrosugars ranged from 0.1 to 0.3, indicating anhydrosugars have degraded during LRT.

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1. Introduction

Aerosol discharged by large-scale biomass burning (BB) activities can lead to regional (Lin et al., 2013), inter-continental (Warneke et al., 2009), and global environmental issues (IPCC, 2013), resulting from direct and indirect effects of aerosols on solar radiation and subsequent climate change (Lin et al., 2014). The direct effect of aerosol components includes absorption of solar energy and warming of the atmosphere by elemental carbon (EC) (Ackerman et al., 2000) as well as scattering of incoming solar radiation back to space by sulfate ion (SO_4^{2-}) (Marmer et al., 2007) and other aerosol constituents, while the indirect effect of aerosol components is through the influence on formation and properties of clouds by acting as cloud condensation nuclei (CCN). To understand CCN formation potential, primary aerosol properties and the precursor gases need to be investigated at the emission sources. Nonetheless, CCN production is enhanced by hygroscopic growth of organic (Kanakidou et al., 2005) or inorganic aerosols (Mochida et al., 2010). Aerosol inorganic species have been known to absorb water effectively under favorable conditions (Lee and Hsu, 2000; Lee and Chang, 2002). Among aerosol inorganic components, SO_4^{2-} is the most abundant component in the great majority of environments. Quinn et al. (2006) found that SO_4^{2-} in total sulfur ($SO_4^{2-} + SO_2$) would increase and the mass fraction of organic matter would decrease after air masses moved from source to downwind areas. Lee et al. (2011) also found that nitrate ion (NO_3^-) levels during the BB period could reach 6.7 and 9.7 fold enhancements relative to the non-biomass burning (NBB) period and a pristine area, respectively. Moreover, microscopic measurements revealed that KCl was predominant in fresh smoke but was converted to K_2SO_4 and KNO_3 in aged smoke (Li et al., 2003). Liu et al. (2000) found excess sulfur formed by condensation of gaseous sulfur compounds on pre-existing potassium ion (K^+)-containing particles during atmospheric transport. Thus, aerosol components will be oxidized during transport from the emission sources to distant locations (Vakkari et al., 2014).

Organic species are abundant in BB aerosols as well (Akagi et al., 2011). The compounds comprising organic aerosol are complex and will experience a series of complicated reactions in the atmosphere. Similar to K^+, levoglucosan is an important tracer of BB aerosols (Simoneit et al., 1999). Fraser and Lakshmanan (2000) showed no degradation of levoglucosan over a period of 10 days under acidic environment. Nevertheless, Gao et al. (2003) suspected that levoglucosan discharged from BB sources may be converted to organic acids during transport under certain conditions. Holmes and Petrucci (2007) suggested that levoglucosan is possibly a precursor of humic-like substances, through acid-catalyzed, OH oxidation, or photo-oxidation processes. Hoffmann et al. (2010) also found levoglucosan may be oxidized by OH radical especially under high relative humidity conditions. Low-molecular-weight dicarboxylic acids and their salts (DCAS) are important in organic BB aerosols (Yang et al., 2013). Among DCAS, oxalic acid and its salts (C2 di-acid) are often the most abundant species. C2 di-acid as a group is not only a major primary component of BB aerosols but also a secondary product (Yang et al., 2009). Different chemical reaction pathways, including in-cloud processing, have been proposed for C2 di-acid production (Kawamura and Yasui, 2005). In addition to C2 di-acid, Kawamura et al. (1996) suggested that malonic acid is produced by photochemical oxidation of succinic acid in the atmosphere. Therefore, the malonic acid over succinic acid ratio can be applied to evaluate the degree of photochemical oxidation during aging of aerosols (Kawamura and Yasui, 2005). For such an application, Aggarwal and Kawamura (2008) compared malonic acid over succinic acid ratios for several locations and found the ratios in a city (Sapporo, 0.88–2.4 with a mean of 1.4) were lower than those of marine aerosols (Chichi-jima Island, 2.0) and remote areas (Pacific, 3.9).

Since the hygroscopic nature of organic aerosols is important for the investigation of CCN formation potential, water-soluble organic carbon (WSOC) is often determined for BB and other aerosols. The group of WSOC is a complex mixture of aromatic and aliphatic compounds (e.g., Zhang et al., 2012). Lim et al. (2010) have shown that secondary organic aerosol (SOA) is readily formed upon oxidation by OH radical in aqueous media. Since WSOC is a major constituent of SOA, production of SOA and subsequent aging of OC results in increase of WSOC (Aggarwal and Kawamura, 2009). Similarly, the production of SOA also increases OC/EC ratios for aged particles observed at background and rural areas compared to urban locations (Aggarwal and Kawamura, 2009). In contrast to WSOC, less water-soluble OC has been considered to be mainly associated with primary emissions from biomass/biofuel burning (Miyazaki et al., 2009) and fossil fuel combustion.

Given the fact that tremendous amounts of BB aerosols originating from Southeast Asia can be transported to southern China (Deng et al., 2008) and Taiwan (Sheu et al., 2010; Lee et al., 2011; Lin et al., 2014) and even further to North America (Peltier et al., 2008), a method to assess mass fraction changes of BB aerosol components (referred as modification throughout the whole paper) during long-range transport (LRT) can help to understand the source-path-receptor relationship for an aerosol component and its environmental impact. In this study, aerosol particles were collected at hill tops in Chiang Mai province (upwind) in Thailand, where BB activities were densely distributed in the surroundings of the sampling site, and at Mt. Lulin (downwind) in Taiwan, where BB aerosols would pass over under prevailing westerlies from Indochina during spring. Aerosol components such as water-soluble inorganic ions (WSII), carbonaceous content, water-soluble/insoluble organic carbon (WSOC/WIOC), DCAS including C2 di-acid, malonic acid and salts (C3 di-acid), succinic acid and salts (C4 di-acid), and glutaric acid and salts (C5 di-acid), and anhydrosugars (including levoglucosan, mannosan, and galactosan) were measured at both locations. Through comparisons of aerosol properties at the upwind and downwind sites, this study proposes the utilization of a computed factor to assess aerosol modifications during transport from the Indochina BB source region over a transport distance of 2400 km.

2. Methods

2.1. Sampling site and sample collection

Fig. 1 shows the geographic location of the observation sites. The Suthep (98° 53′E, 18° 48′N, 1396 m a.s.l.) and Doi Ang Khang (DAK) sites (99° 05′E, 19° 93′N, 1536 m a.s.l.) are both located in the Chiang Mai mountains in northern Thailand. The distance between the Suthep and DAK sites is about 190 km with very similar environments at both sites. Densely distributed fires were visible at both sites and detected from satellites. The Lulin Atmospheric Background Station (Mt. Lulin) (120° 52′ 25″ E, 23° 28′ 07″ N, 2862 m a.s.l.) is located on the summit of Mt. Lulin in central Taiwan where there is unobstructed view over the surrounding mountains. The rising of BB pollutants from surface to the atmosphere over northern Indochina and southern China and further transport to the Western Pacific under the prevailing westerlies has already been shown in recent studies (Lin et al., 2013, 2014; Yen et al., 2013). Hence, although Chiang Mai and Mt. Lulin are separated by 2400 km, BB pollutants originating from northern Thailand can still be transported in the upper atmosphere over Taiwan. This phenomenon is discussed in more details in sections 2.3 and 3.1.

The study period for the Mt. Lulin site was from 12 March to 31