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Sources and formation pathways of organic aerosol in a subtropical metropolis during summer

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HIGHLIGHTS

- Biogenic contributions to OA formation are strong in both urban and rural Taipei.
- SOA dominated OA production, and its fraction increases with height in the PBL.
- Major contribution of OA from aqueous-phase processes.
- PBL height & sunlight variations and local circulation controlled OA diurnal cycle.

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ABSTRACT

A field campaign combined with numerical simulations was designed to better understand the emission sources and formation processes of organic aerosols (OA) in a subtropical environment. The field campaign measured total and water soluble organic carbon (OC) in aerosol, as well as its precursor gases in the Taipei metropolis and a nearby rural forest during the summer of 2011. A regional air-quality model modified with an additional secondary organic aerosol (SOA) formation pathway was used to decipher the observed variations in OA, with focus on various formation pathways and the relative contributions from anthropogenic and biogenic sources.

According to the simulations, biogenic sources contributed to 60% and 72% of total OA production at the NTU (urban) and HL (rural) sites. The simulated fractions of SOA in total OA were 67% and 79% near the surface of NTU and HL, respectively, and these fractions increased with height and reach over 90% at the 1-km altitude. Estimated from the simulation results, aqueous-phase dicarbonyl uptake was responsible of 51% of OA production in the urban area, while the primary emissions, reversible partitioning of semi-volatile oxidation products, oligomerization of semi-volatile SOA in the particulate phase and acid-enhanced oxidation contributed to 33%, 10%, 5% and 1% respectively; in the rural area, the percentages were 59%, 21%, 13%, 7% and 1%, respectively. Meteorological factors, including large-scale wind direction, local circulation and planetary boundary layer height, all have strong influences on the source contributions and diurnal variations of OA concentration.

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1. Introduction

Organic compounds are one of most abundant components of fine aerosol particles, particularly in areas with substantial fossil fuel and bio-fuel burning (Kanakidou et al., 2005; Zhang et al., 2007). The organic component, which we termed the organic aerosols (OA), can be classified into primary OA (POA) which are directly emitted species, and the secondary OA (SOA) which are formed by chemical reactions in the atmosphere. The fraction of SOA in total organic matter is typically 20–80% of the measured

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mass (Carlton et al., 2009; He et al., 2011), but the values have large spatial and temporal variations. Kanakidou et al. (2005) reported that 20–50% of the total fine aerosol mass in the mid-latitudes is organics, and the fraction can be up to 90% in tropical forested areas, indicating the importance of biogenic sources in warm and humid environments.

Although field measurements may provide the spatial and temporal distributions of OA compositions, it is difficult to decipher the sources and formation processes of OA due to their complex chemical mechanisms and variation with meteorological conditions. A more comprehensive way for understanding OA production pathways is to combine the advantages of measurements and numerical models with resolved chemical and meteorological processes. Because of the complexity of precursor volatile organic compounds (VOCs) and the unclear formation pathways of SOA, including gas-to-particle conversion and heterogeneous chemistry (Gard et al., 1998; Kanakidou et al., 2005), simulating the SOA cycle using air quality models remains a challenging task (Hallquist et al., 2009). In traditional models, SOA forms via the partitioning of precursors' oxidation products onto the existing organic aerosol (Odum et al., 1996; Pankow, 1994). More recent models have considered SOA formation by in-cloud reactions (Carlton et al., 2008). However, the simulated SOA in the boundary layer and aloft usually remain underestimated (Carlton et al., 2010; Heald et al., 2011; Jathar et al., 2014), so some SOA formation processes may be missing in the models.

Taipei is a major metropolitan area surrounded by mountains covered in subtropical rainforests, so emission of SOA and its precursors are ample from both anthropogenic and natural sources. A few studies have reported measurements of carbonaceous aerosol concentrations in Taiwan. For example, Chou et al. (2010) found that POA and SOA each contributed 11 ± 6% and 18 ± 4%, respectively, of the total $PM_{2.5}$ in Taipei, according to measurements conducted in 2003–2007. However, detailed analysis of source contributions and formation processes associated with carbonaceous aerosols, particularly SOA, are scarce in this area.

To understand the impacts of human activities on the physicochemical properties of aerosols over such an urban area, a field campaign was conducted over the Taipei metropolis and an adjacent rural area in summer 2011. The US EPA Models-3/Community Multi-scale Air Quality (CMAQ) model was also applied to simulate organic aerosol formation during this field campaign to investigate its various sources and formation pathways. The model was modified following Li et al. (2013) by considering the irreversible uptake of dicarbonyls by aqueous particles as an additional pathway for SOA formation. The simulations results were evaluated by comparison with data collected during the field campaign. The discussions focus on the model performance with respect to carbonaceous compounds in the Taipei metropolis and the contributions of various sources and formation processes to OA concentrations.

2. Methodology

2.1. Measurements

The Taipei metropolis is a densely populated region with substantial anthropogenic emissions. It is located in a subtropical basin surrounded by highly vegetated mountains or highland (see Fig. 1a), and thus is a unique location for studying the complex contributions of OA from anthropogenic versus biogenic sources. For understanding the characteristics of OA in Taipei and the adjacent areas, a field campaign conducted by the Academia Sinica and National Taiwan University (hereafter referred to as NTU) was conducted during Aug. 15–21, 2011. Two contrasting measurement sites were selected, one at the urban area and the other in a rural area (see location map in Fig. 1a). The NTU campus site (25.02°N, 121.53°E; black star in Fig. 1a) is located in the southern part of the city. The sampling inlet was set up on the top floor of a building, and is approximately 25 m above ground. The HuaLin site (hereafter referred to as HL; 24.88°N, 121.56°E; yellow star in Fig. 1a) is in a rural environment, located 15 km south of NTU and 10 km away from the nearest suburban city of Sindian. At an elevation of 410 m, the HL site is surrounded by bushes and broad-leaved trees, and without significant human activities.

Simultaneous chemical measurements at NTU and HL include isoprene, O_3, elemental carbon (EC), and organic carbon (OC) mass concentrations, the latter two were measured with Sunset semi-continuous OC-EC Field Analyzer (Sunset OC-EC). Two sets of 12-hr-accumulation filter samples were collected daily at each site, one from 8 a.m. to 8 p.m. local time to represent daytime conditions, and the other from 8 p.m. to 8 a.m. to represent nighttime conditions. The water-soluble organic carbon (WSOC) concentration was computed as the difference between water-soluble total carbon (WSTC) and water-soluble inorganic carbon (WSIC, is the structural basis for inorganic compounds such as gas carbonates

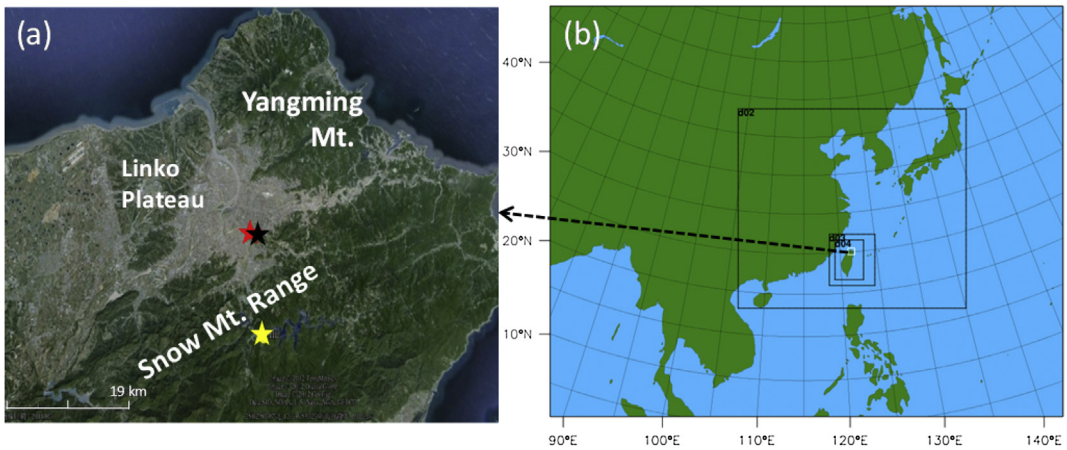

Fig. 1. (a) Satellite image of the Taipei metropolis (gray shading area) and its surroundings: the Yangming Mt. (peak elevation 1120 m) to the north, the Snow Mt. Range (peak elevation 3703 m) that extends from the east to the southwest, and the Linko Plateau (elevation 250 m) to the west. The locations of NTU, HL and TWEPA Guting sites are indicated by the black, yellow and red stars, respectively. (b) Geographic view of the model domains for the simulations in this study, and the white-rectangle area corresponds to the domain in (a). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

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