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Anhydrosugar characteristics in biomass smoke aerosol—case study of environmental influence on particle-size of rice straw burning aerosol

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ABSTRACT

Burning of agricultural residues, such as rice straw, is a sizeable source of smoke aerosol particularly in Asia. During a field study on the subtropical island of Taiwan ambient aerosol was examined for seasonal patterns in terms of size-resolved chemical composition. Ambient concentrations, size distributions and diagnostic ratios of three anhydrosugars (levoglucosan, mannosan, and galactosan), used as molecular tracers for biomass burning, are reported here. Levoglucosan concentrations showed a strong seasonal dependence in the ambient levels as well as size distributions. During the wet (summer monsoon) season levoglucosan was observed with average concentrations of 437, 882 and 1116 ng m^{-3} in $PM_{2.5}$, PM_{10} and TSP, respectively. In contrast, winter/dry season levoglucosan levels were lower with 436, 448, and 559 ng m^{-3} on average in $PM_{2.5}$, PM_{10} and TSP, respectively. The particle-size distributions were characterized by a drastically increased coarse ($PM_{2.5-10}$) fraction in the summer season, which was 36 times higher than during winter, likely due to higher fuel moisture content as well as enhanced condensation, coagulation and hygroscopic growth of the relatively fresh smoke particles derived from rice straw burning. Furthermore, the relative abundance of the three anhydrosugars was examined for characteristic signatures in respect to different types of biomass.

1. Introduction

Biomass combustion processes release substantial amounts of pollutants into the atmosphere on a global basis. South, Southeast and East Asia in particular are major source regions of smoke particles derived from biomass and biofuel burning (Streets et al., 2003). The principal combustion processes include open burning of biomass, either in form of agricultural residue burning (anthropogenic) or wildland (such as forests or Savanna, i.e., natural) fires, as well as domestic use of biofuels for cooking and heating (anthropogenic). Combustion of agricultural waste material, such as straw and stubble, constitutes a particularly large source of atmospheric particulate matter, and especially carbonaceous aerosol, throughout Asia (Gadde et al., 2009; Yevich & Logan, 2003). It is well known that aerosol particles in general and smoke aerosol in particular exert significant impacts on the environment, including global climate, regional air quality, and

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human health (Mauderly & Chow, 2008; Poeschl, 2005; Reisen et al., 2011). Yet to date, the understanding of aerosol source processes, their atmospheric transformations and their chemical as well as physical characteristics is still rather limited. Specifically, the chemical composition of organic aerosol components as a function of particle size has been investigated only for certain compounds and locations (e.g., Alves et al., 2000; Herckes et al., 2006; Kavouras & Stephanou, 2002; Rissler et al., 2006; Wang et al., 2011a, b; Yang et al., 2006), while size-dependent aerosol properties are critical in terms of their effect on many environmental processes, such as human health, interaction with incident light and formation of cloud droplets.

Molecular tracer techniques have been applied in several investigations to assess the contributions of biomass burning smoke to the ambient aerosol burden (e.g., Caseiro et al., 2009; Engling et al., 2006b; Holden et al., 2011; Kourtchev et al., 2011; Reisen et al., 2011; Ward, Rinehart & Lange, 2006; Zhang et al., 2010). Particularly, the anhydrosugars levoglucosan, mannosan, and galactosan, derived from the thermal breakdown of the plant building blocks cellulose and hemicellulose, are good source tracers for biomass or biofuel burning due to their source-specific generation and atmospheric stability (Fraser & Lakshmanan, 2000; Shafizadeh et al., 1979; Simoneit et al., 1999), although caution is warranted in the use of these tracers for quantitative assessments of source contributions, especially in cases when the smoke aerosol may have encountered water (e.g., in form of fog or clouds) and strong oxidants during atmospheric transport (Hennigan et al., 2010; Hoffmann et al., 2010). Nevertheless, these anhydrosugars can provide valuable information about the influence of biomass burning activities on a qualitative level or allow for conservative quantitative estimates of source contributions, as shown by long-term investigations of levoglucosan concentrations upon long-range transport to a site in the western North Pacific Ocean (Mochida et al., 2010). In addition, diagnostic ratios of certain anhydrosugars can provide insights into the specific types of biomass which were burned (Engling et al., 2009; Fabbri et al., 2009).

Whereas in Europe and North America wood combustion is most prevalent, agricultural burning activities, e.g., in form of post-harvest burning of crop residues, are major sources of biomass smoke in Asia and Taiwan in particular (Cheng et al., 2009; Lee et al., 2008). Furthermore, ambient conditions in Taiwan differ significantly from those in regions with moderate climate, especially in terms of relative humidity and amount of rainfall. In a recent study by Lee et al. (2008), it was observed that atmospheric conditions may influence the characteristics of smoke particles, specifically in terms of their particle-size dependent chemical composition. Therefore, the aim of this study was to demonstrate the influence of ambient atmospheric conditions, in form of seasonal patterns, on the size-dependent chemical characteristics of ambient aerosol particles, influenced by biomass burning emissions.

2. Experimental

2.1. Aerosol sample collection

The primary agricultural commodity in Taiwan is rice, similar to most countries in Asia. Yunlin County is one of the major rice growing regions in Taiwan, contributing 17% of Taiwan's total rice production (www.afa.gov.tw/GrainStatistics). Generally, the rice production in Taiwan is divided into two growing periods, with the summer harvest providing nearly twice the rice yield compared to the winter harvest. Ambient aerosol samples were collected between December 2006 and August 2007 in Yunlin County, located in south-central Taiwan, on the campus of the National Yunlin University of Science & Technology in Douliou City (23° 41′ 40.2″N, 120° 32′ 10.9″E). The entire study was divided into three periods: (1) winter harvest season, (2) background period (spring), and (3) summer harvest season. Separate day and night time samples were collected during the two harvest seasons, with typical sampling times of 12 h, whereas 24 h sampling was conducted during the background period (spring 2007). The daytime sampling period during the summer season was abridged by up to 50% on certain days due to rainfall in the afternoon (the average summer season precipitation in Douliou was ~4.5 mm/day). In total, 32 samples were collected, including 9 samples during the winter and 13 during the summer harvest season, and 10 samples were obtained during spring to represent the background period. The samples collected during winter were also part of another recent study conducted by Lee et al. (2008).

The aerosol samples were collected with several high-volume (Hi-vol) air samplers and one medium-volume (med-vol) sampler, consistently during the 3 measurement periods. The med-vol sampler (UAS Model M310; MSP Incorp., Shoreview, MN, USA) was employed to collect $PM_{2.5}$ and $PM_{2.5-10}$ particles, while a PM_{10} and a TSP Hi-vol sampler (Thermo Andersen, Franklin, MA, USA) were operated simultaneously to collect particles with aerodynamic diameters of ≤ 10 μm and total suspended particulate matter (TSP), respectively. The $PM_{>10}$ fractions were calculated by subtracting PM_{10} from TSP concentrations. The sampling precision for the size-resolved samplers (UAS and PM_{10} Hi-vol) was verified to be within 10%. Samples were collected on pre-fired (at 500 °C) quartz fiber filters (Tissuquartz, 2500 QAT-UP; Pall Corporation, East Hills, NY, USA) for anhydrosugar measurement and on glass fiber filters (Type A/E; Pall Corporation, East Hills, NY, USA) for PM mass determination. All sample filters were stored at −20 °C until sample analysis. Quality control procedures also included collection and analysis of three field blank samples during each season. A weather sensor (Vantage Pro II, Davis Instruments, Hayward, CA, USA), co-located with the aerosol samplers, was used for collection of meteorological parameters.

The meteorological conditions during the different seasons were characterized by fairly high temperatures, ranging from 18.3 ± 1.2 during the winter harvest season to 30.5 ± 0.9 °C in the summer, and low to medium wind speeds (with a range of 2.9 ± 1.4 in spring to 4.0 ± 3.1 m s^{-1} during the winter harvest season), predominantly northerly and