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Stable carbon fractionation in size-segregated aerosol particles produced by controlled biomass burning

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ABSTRACT

Six different biomass fuel types (wood pellets, sunflower stalk pellets, straw pellets, buckwheat shells, mixed biomass waste pellets, and grain screenings) and wastewater sludge pellets were burned under controlled conditions to determine the effect of the biomass type on the emitted particulate matter mass and stable carbon isotope composition of bulk and size-segregated particles. Aerosol particles were sampled using the total suspended particle (TSP) sampler and a micro-orifice uniform deposit impactor (MOUDI). The results demonstrated that particle emissions were dominated by the submicron particles (size < 1 μm) in all biomass types. However, significant differences in emissions of submicron particles and their dominant sizes were found between different biomass fuels. The isotopic fractionation between aerosol particles and original biomass material varied from -0.94 ± 0.23‰ to 1.12 ± 0.16‰. The largest negative fractionation -0.94 ± 0.23‰ was obtained for the wood pellet fuel type while the largest positive isotopic fractionation (1.12 ± 0.16‰) was observed during the grain screenings combustion. The carbon isotope composition of MOUDI samples compared very well with the isotope composition of TSP samples indicating consistency of the results. The measurements of the stable carbon isotope ratio in size-segregated aerosol particles suggested that combustion processes could strongly affect isotopic fractionation in aerosol particles of different sizes thereby potentially affecting an interpretation of ambient atmospheric observations.

1. Introduction

Biomass burning is the largest source of primary fine fraction carbonaceous particles. Also it is the second largest source of trace gases in the global atmosphere (Andreae & Merlet, 2001; Bond et al., 2004). Subsequently biomass burning has a strong effect not only on the regional scale but also in areas distant from the source (Alves et al., 2011; McMeeking et al.,

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2009; Ryu et al., 2007). Overall, 95% of the global biomass burning is anthropogenic resulting from burning of woodland, pastures and agricultural waste after harvesting, residential biomass fuel combustion and cooking stoves in developing countries (Andreae & Merlet, 2001; Crutzen & Andreae, 1990). During winter, biomass combustion is one of the major sources of organic aerosol over large parts of Europe (Puxbaum et al., 2007). The variability in properties of biomass fuels is huge and may significantly impact air quality during combustion processes. Therefore, it is important to determine environmentally friendly fuels and fuel mixtures to minimize particle and flue gases emissions. There are a limited number of studies on the effects of combustion of various alternative biofuels (e.g. crops, wood bark or agricultural byproducts) on physico-chemical properties of fine fraction particles. Moreover, the biomass burning can contribute almost half of the organic aerosol mass in the area affected by wildfires, wood or waste burning (Dusek et al., 2013; Kostenidou et al., 2013; Mollicone et al., 2006; Ortiz de Zarate et al., 2005; Zauscher et al., 2013).

Emissions of particles or gases during biomass burning heavily depend on the combustion conditions, which are either flaming, in which a more complete oxidation is occurring, or smoldering often referred as an incomplete combustion (Koppmann et al., 2005). A characterization of these two phases is required when biomass burning emissions are estimated. Reid et al. (2005) reported that aerosol particles generated by biomass burning consisted mainly of carbonaceous compounds (mostly organic carbon (OC), and small amounts of black carbon (BC)) and small amount of various inorganic components. This inorganic fraction mostly consists of insoluble dust, ash, soluble salts (potassium/ammonium sulfate) and acids (Janhäll et al., 2010; Reid et al., 2005). Most of the particles emitted during the biomass burning are in the fine fraction (Badarinath et al., 2009), with a count median diameter of 100–150 nm often referred to as accumulation mode. A coarse fraction consists of dust, carbon aggregates, ash and residual ash (Formenti et al., 2003; Hungershoefer et al., 2008). Sometimes a nucleation mode is present as well (Janhäll et al., 2010; Radke et al., 1991).

The stable isotope method is a powerful tool for source identification and can be combined with other source apportionment techniques. Therefore, this method can be used to study biomass burning events. Fu et al. (2012) demonstrated that crop-residue burning activities can significantly enhance the organic aerosol loading and alter the organic composition and stable carbon isotopic composition of aerosol particles in the troposphere over the North China Plain. Ulevicius et al. (2010) showed that a major part of carbon mass in aerosol particles transferred by the continental air masses from the wildfire location was from vegetation burning. The application of isotope techniques to investigate combustion related aerosol sources requires knowledge of the isotope values of the original biomass, as well as the associated isotope fractionation effects of combustion derived components. The stable carbon isotope ratio ($^{13}C/^{12}C$) elucidates carbon emissions associated with different plants, both terrestrial and oceanic, due to preferential photosynthesis uptake routes of heavier or lighter inorganic carbon isotopes (Ceburnis et al., 2011; Huang et al., 2010). The carbonaceous compounds emitted from C_3 plants have a distinctly different isotope signature ($\delta^{13}C$ ranging from $-20.0‰$ to $-32.0‰$) from those of C_4 plants ($-9.0‰$ to $-17.0‰$) (O'Leary, 1981). In addition, isotopic variability depends on the chemical make-up of the plant, with the lipid fraction being depleted in ^{13}C by typically 5‰ relative to the bulk plant material (Deines, 1980; Kennicutt et al., 1992). Carbon isotopic fractionation during cellulose and lignin synthesis imparts an isotopic difference relative to the whole wood of approximately 1‰ and $-2‰$, respectively (Loader et al., 2003).

Many studies have often assumed no significant carbon isotope fractionation occurring between black carbon and the original vegetation during combustion (Bird & Gröcke, 1997; Leavitt et al., 1982; Schleser et al., 1999). However, other studies suggested that stable carbon isotope ratios of char or BC may not reliably reflect carbon isotopic signatures of the source vegetation. For example, Cachier et al. (1985) reported that aerosol derived from combusting C_4 grass was depleted in ^{13}C by up to 7.0‰ compared to the original grass, whereas aerosol derived from C_3 vegetation was enriched in ^{13}C by 2.0‰. Krull et al. (2003) observed a ^{13}C depletion of up to 8‰ in C_4-derived chars from wild combustion, but there was no significant isotopic change in chars from wood or C_3 grasses. The isotopic fractionation during combustion has also been studied by Turekian et al. (1998), who reported positive fractionation (by 0.5‰) in aerosol particles compared to the source vegetation, formed during burning of C_3 plants. They also observed a ^{13}C depletion (by 3.5‰) in aerosol particles formed during burning of C_4 vegetation relative to the source plant material. Overall, the apparently conflicting results throughout the literature regarding the observed fractionation suggest that combustion conditions may be responsible for the observed effects.

The purpose of the present study was to gather more quantitative information on carbonaceous aerosols produced in controlled biomass burning, thereby having a potential impact on interpreting ambient atmospheric observations. Our objective was to better quantify the fractionation of carbon isotopes between the original biomass and aerosol particles emitted during the controlled combustion and its potential dependence on aerosol particle sizes for several biomass types.

2. Materials and methods

2.1. Types of biomass fuels

A total of six different types of biomass fuels were investigated during the experiment. In addition to wood pellets that are one of the most popular biomass fuels for the pellet burner, sunflower (*Helianthus annuus*) stalk pellets, straw (*Triticum aestivum*) pellets, buckwheat (*Fagopyrum esculentum*) shell and grain screenings were investigated (Table 1). Wastewater sludge pellets were also used as biofuel. Straw pellets are commercially available in Lithuania and widely used in residential heating. Grain screenings are used in industry while wastewater sludge pellets are rarely used as biofuels. All of biomass burning materials were C_3 plants except wastewater sludge. The exact origin and composition of the wood and mixed