



Full Length Article

Time-resolved chemical composition of small-scale batch combustion emissions from various wood species



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ABSTRACT

Small-scale batch combustion of wood is a major source of fine particles, black carbon emission and polycyclic aromatic hydrocarbons in Finland. The mass and chemical compositions of batch combustion emissions are known to be highly time-dependent. In this study, the gaseous and particulate batch combustion emissions of three European wood species (beech, birch and spruce) were quantified in detail with an extensive set of online analysers, including a soot particle aerosol mass spectrometer (SP-AMS) for real-time detection of particulate chemical composition. Ignition and a new batch addition on top of glowing embers were identified as the primary low temperature events during which both particulate and gaseous organic emissions peaked. The flaming combustion created high temperature conditions and produced increased emissions of refractory black carbon (rBC) and PAHs. The residual char combustion phase was characterized by low particulate mass emission consisting mainly of alkali salts and elevated concentrations of gaseous organic emissions and CO. Overall, hardwood species (beech and birch) had the highest PM₁ emissions, and the difference between the lowest average emission (spruce) and the highest (birch) was more than 3-fold. The increasing combustion chamber temperature during sequential combustion of wood batches was found to decrease the carbonaceous fraction of the PM as well as OC/EC ratio, as the result of more efficient secondary combustion.

1. Introduction

Small-scale batch combustion is known to emit relatively high amounts of particulate and gaseous emissions compared to, for example, light fuel oil combustion or continuous combustion of wood [1–3]. In Finland, small-scale wood combustion is the second largest heat source in residential houses, after district heating [4], producing a major fraction of the fine particles, black carbon emissions (BC) [5] and polycyclic aromatic hydrocarbons (PAH) [6]. Elevated particulate matter (PM) levels are problematic throughout Europe, especially in dense urban areas in wintertime when the demand for heating is increased and the dispersion of emissions may be decreased due to temperature inversion [7]. The dispersion of small-scale combustion emissions is generally weak compared to large or medium scale energy production due to the low emission height. In addition, the BC

emissions, which are deposited in arctic areas, lead to a decrease of the snowpack albedo, heating of the snow and acceleration of its melting in springtime [8].

The nature of the batch combustion process is highly dynamic and emissions have considerable variation according to the phase of burning [9–11]. To evaluate possible adverse health and climate effects, and developmental needs in batch combustion technology, detailed information is needed on the temporal variation of emissions and possible influence of wood species upon it. Considering the health effects of wood combustion emissions, elemental carbon, PAHs and transition metals, such as zinc, have especially been proposed as potential chemical components inducing toxicological cell responses [12,13]. In addition to the mass of particles, also the particle surface area and size have been proposed to be important factors for the cell toxicity [13,14].

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To date, there are notably few studies reporting the time-dependency of physico-chemical properties of particulate emissions from batch-wise wood combustion [9–11,15]. In those studies, particularly the effect of combustion conditions of wood logs in stoves on the properties of particulate emissions has been described. However, it is not well known how different wood species affect the time-resolved emission properties of logwood combustion. In this study, time-resolved particulate and gaseous emissions from small-scale combustion of different wood species in heat-retaining masonry heater were investigated. These data would broaden the scientific knowledge needed in the research of climate and health effects of small-scale combustion but also in the development of more efficient combustion devices as well as secondary emission reduction technologies.

From the particulate emissions under observation were time series of the number concentration, number-size distribution, surface area, mass emission and chemical composition (refractory black carbon, organics, K^+ , Cl^- , NO_3^- , SO_4^{2-}). Also the time series of selected PAH compounds and the relative emission of zinc were the subjects of specific interest because of their adverse effects on human health. The gaseous components oxygen (O_2), carbon dioxide (CO_2), carbon monoxide (CO), organic gaseous compounds (OGC) and nitrogen oxides (NO_x) were monitored in real time. Furthermore, particle samples were collected on filters for chemical and gravimetric analysis. In addition to time-series, the emission factors were calculated for the main emissions. This kind of detailed analysis also provides an opportunity to study whether any correlation exists between single gas components (e.g., CO or OGC) and, for example, PAHs.

2. Materials and methods

In these experiments, three different wood species were combusted in a modern masonry heater (MMH). The combustion experiments were carried out at the Fine Particle and Aerosol Technology Laboratory in the University of Eastern Finland.

2.1. Combustion appliance, fuels & procedure

The batch combustion appliance used in this study represents a modern type of masonry heater with combustion air staging and a heat retaining structure. The high mass of soapstone (~1300 kg) acts as a heat reservoir. The heater had an enclosed upright firebox with a ceramic lining and a double window glass door. Furthermore, the heater featured a special rack (Fig. S1, Supporting Information) placed in the firebox, where the logs were disposed according to the instructions given by the stove manufacturer (described in detail below). Like in the majority of heat retaining fireplaces the flue structure was contra flow, where flue gases flow from the upper combustion chamber downward to side ducts and then into the stack. The stack was placed below a hood connected to a flue gas fan, to produce an unforced draft. The draft was regulated with two dampers. The target value for pressure in the stack was 12.0 ± 0.5 Pa below ambient pressure. The pressure was reduced by closing the damper a little whenever the pressure exceeded the target value. The combustion air was distributed between primary airflow through the grate, secondary airflow above the fuel batch and window flushing air. A schematic drawing of the combustion chamber can be found as Supporting Information (Fig. S2). Proportions from the total combustion air were approximately 20%, 45% and 35%, respectively (these values are reported by the heater manufacturer and represent average values). Air-staging in batch combustion enhances the mixing of pyrolysis and gasification gases and air, which leads to more complete combustion during the flaming phase and thus reduces gaseous emissions (CO , OGC) and particulate organic emissions compared to conventional non-staged combustion [16].

The combustion experiments were carried out with three different wood species, European beech (*Fagus sylvatica*) (6 experiments), birch (*Betula pendula*, *Betula pubescens*) (7 experiments) and Norway spruce

(*Picea abies*) (4 experiments). Fuel samples were analysed for the contents of ash, moisture, calorific value, as well as the elements: C, H, O, N, S, Cl, Cr, Cu, K, Na, and Zn.

Each experiment lasted for four hours, during which six batches of wood were combusted. In the 1st batch, ten logs were laid crosswise (0.23 kg each) and on top of them were 150 g of smaller wood pieces as kindling. Ignition of the 1st batch was always done from the top. Batches 2–6 consisted of five logs 0.5 ± 0.05 kg each. The weight of each batch was 2.50 ± 0.02 kg, so in each experiment the total amount of wood combusted was 15.0 ± 0.1 kg.

The combustion procedure was made as consistent as possible for all the wood species by conducting a series of pre-tests. However, due to differences in properties between the wood species (moisture, density, composition), minor alterations were made to the procedure depending on the wood species. The duration of the 1st batch of the hardwood species was 5 min shorter than with spruce. Thus, with beech and birch the 1st batch lasted for 30 min, while with spruce the duration was 35 min. In all cases, batches 2–6 were combusted for 35 min and after the sixth batch, no more wood was introduced, but the embers were stoked and the secondary air channels were closed according to the instructions given by the stove manufacturer. The char burning phase went on until four hours was full (35 min with beech and birch, 30 min with spruce), after which the experiment was finished. Because the combustion experiments occurred on consecutive days, the cooling of the combustion appliance was enhanced with a blower between the experiments, in order to cool the stove down to the room temperature. The flue gas temperature was measured from the stove flue gas outlet with a K-type thermocouple.

2.2. Gaseous emissions

Concentrations of carbon monoxide, carbon dioxide, oxides of nitrogen, organic gaseous compounds and oxygen were measured from the flue gas through an insulated and externally heated sampling line ($180^\circ C$) (Fig. 1) with single component gas analysers (Magnos 106, Uras 14 and Limas 11 UV; ABB). The OGC concentrations were measured with a flame ionization detector (Multi-FID 14, ABB), calibrated against propane. CO and NO_x concentrations were measured at ppm level, the OGC was measured at the level of $mgC\ m^{-3}$, while O_2 and CO_2 were measured at the accuracy of 0.01%.

2.3. Particle sampling and dilution

For the online particle analysers, dilution of the sample was required because of the high concentrations of particulate matter and water, and high temperatures in the raw flue gas. The particle samples were taken with the combination of a porous tube diluter (PRD) and an ejector diluter (ED) [17,18]. Dilution air was purified with a zero air generator (737-15, AADCO Instruments Inc). The dilution ratio (DR) after the PRD-ED combination was adjusted to 1:40 based on the CO_2 concentrations in raw flue gas and in the diluted sample [18]. An additional ejector diluter, which had a fixed DR of 1:10, was installed to decrease the mass concentrations to optimal measurement range for the most sensitive online analysers, and so the total DR for those devices was approximately 1:400 (Fig. 1).

Particle number size distribution was measured from diluted flue gas with an Electric Low Pressure Impactor (ELPI, Dekati) (DR 1:40) and a Scanning Mobility Particle Sizer Spectrometer (SMPS, TSI) (DR 1:400). The ELPI was operated with a 10 lpm sample flow and a filter stage. Collection plates of the ELPI were sintered and vacuum oil-embedded. The SMPS featured a Differential Mobility Analyzer (DMA 3081, TSI) and a Condensation Particle Counter (CPC 3775, TSI) at a flow rate of 0.3 lpm. The surface area of the particulate matter was measured with a Nanoparticle Surface Area Monitor (NSAM 3550, TSI) (DR 1:400). The NSAM was used to measure the total particle surface area by utilizing the smallest possible trap voltage without allowing gas

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