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Aerosol in emissions of Siberian biomass burning: small-scale fire study

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Abstract

A better understanding of aerosol emissions from Siberian boreal forest fires is crucial, as they exert critical environmental and climate impacts in subarctic regions and the Arctic. The ability of biomass burning aerosol to absorb/scatter incoming radiation as well as act as cloud condensation nuclei strongly depends on microphysical, chemical and hygroscopic particle characteristics, for which a comprehensive regional database does not exist to date.

In this work small-scale combustion experiments were performed with the purpose to fill the gaps in available data on particulate emissions from Siberian boreal forest fires. Typical regional biomass species, pine and debris, were burned in a Large Aerosol Chamber (LAC) under controlled combustion conditions representative of wildfires and prescribed burns. Comprehensive physico-chemical characterization of smoke aerosols are performed, including morphology, elemental composition, carbon and ion content, organic/inorganic functionalities, and selected organic compounds.

Individual particle analysis of smoke morphology and elemental composition reveals the strong dependence on combustion temperature, indicating the dominant abundance of soot agglomerates at flaming versus roughly spherical organic particles at smoldering phase. Similarly, smoldering emissions are characterized by very low EC content (0.4% of PM mass) and high OC/EC ratios (62-194), while flaming emissions contain around 10% of EC and have OC/EC ratios in the range 0.32-0.57. Calcium and potassium combined with sulfates, chlorides, phosphates, and nitrates compose inorganic salts found dominantly at flaming while the low temperatures during the smoldering phase are not sufficient for salt formation. Both flaming and smoldering smoke aerosols are characterized by high levoglucosan concentration (up to 25% of PM mass), confirming levoglucosan as a good

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molecular marker for Siberian boreal forest wood burning. The ratio of levoglucosan/mannosan ~ 2-4 is found as an indicator of softwood burning.
This study shows the importance of obtaining the chemical characteristics for dominant PM components in order to enable the assessment of contributions from Siberian biomass burning to atmospheric pollution and the aerosol/climate system.

Keywords: Biomass burning; combustion phase; physico-chemical properties.

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1. Introduction

Biomass burning is known to be a major contributor to the global aerosol budget (Ito & Penner, 2005). It releases into atmosphere particulate species such as black carbon (BC) (mostly absorbing visible light), organic carbon (OC) and fly ash. The direct and indirect impacts of BC aerosols induce the highest uncertainty amongst climate active species with respect to temperature changes, radiative forcing, and cloud formation. At the regional level, wildfire smoke degrades air quality and significantly changes the ambient aerosol properties (Popovicheva et al., 2014). Especially in megacity urban areas, biomass burning may profoundly affect the air quality and public health, indicating smoke aerosol as harmful pollution (Amiridis et al., 2012). Detailed information on wildfire aerosols is crucial for understanding the origin as well as environmental and climate impacts they may have in the atmosphere.

Arctic climate is especially BC-sensitive because of deposition on snow, accelerating icecap melting and changing the surface albedo (Quinn et al., 2011). Wildfires are the primary source of aerosols in the Arctic and sub-Arctic regions during the spring/summer months. Particularly, plumes of Siberian boreal forest fires are often found approaching the Arctic coast at approximately 70^0N (Paris et al., 2009) and are considered a major source of climate-relevant species emitted at northern latitudes. Major organic components in wildfire smoke are monosaccharide anhydrides, derivates from cellulose breakdown. For source apportionment levoglucosan and mannosan are used as molecular markers of wood burning and other types of biomass burning (Engling et al., 2006, 2013). Small-scale studies in combustion chambers may provide useful insight into the characterization of biomass burning emission at controlled conditions (Chen et al., 2007; Engling et al., 2006) as well as determination of the relationship between physical, chemical and optical properties (Hopkins et al., 2007). Despite extensive research on biomass burning emissions, there is considerable uncertainty over chemical speciation and molecular markers of Siberian boreal forest wildfires.

This paper presents a comprehensive study of microscopic, optical, and chemical characteristics of smoke aerosols of boreal forest wildfires with the major purpose to quantify Siberian atmospheric pollution and impacts of combustion source emissions on changes in the aerosol/climate system in subarctic regions and the Arctic. A detailed characterization of physico-chemical properties of biomass burning particulates was performed, including morphology, elemental composition, surface chemistry, ion, elemental (EC) and organic carbon (OC) content, molecular markers, and individual organic compounds. Special attention is given to the identification of morphological and chemical micromarkers of combustion aerosols specific for Siberian boreal wildfires.

2. Experimental

Laboratory simulations of wildfire biomass burning were performed in a Large Aerosol Chamber (LAC) of 1800 m^3 of the Institute of Atmospheric Optics, RAS, in Tomsk city (Siberia). The major sources of smoldering, open flaming, and mixed burning of typical biomass types of central Siberian forests (pine wood and conifer forest debris) are simulated under controlled combustion conditions. Size-segregated PM10 and PM2.5 and BC fraction were studied using gravimetry, polarization spectronephelometry of scattering radiation, and aethalometry. Scanning electron microscopy (SEM) coupled to energy-dispersive X-ray (EDX) spectroscopy was used for characterization of individual particle morphology and elemental composition. It was supported by cluster analysis for the quantification of major groups of smoke particles with the purpose to discriminate between the different types of fire-emitted aerosols. Additionally, we combined measurements from four bulk characterization techniques including thermal-optical analysis, FTIR spectroscopy, liquid and ion chromatography, and GC-MS to determine carbon fractions, organic/inorganic composition, ionic and individual organic compounds.

3. Siberian biomass smoke microstructure

Individual particle analysis of smoke morphology and elemental composition reveals a strong dependence on combustion temperature, indicating the dominant presence of soot agglomerates from flaming burns versus roughly spherical organic particles from the smoldering phase. Cluster analysis of smoke microstructure was able to separate fire-emitted particles into two characteristic groups ("Group Soot" and "Group Organic") which comprise around 90% and 60% of total particle numbers from flaming and smoldering phases, respectively. Their representative SEM micrographs are shown in Fig.1. Apart from C and O, significant quantities of elemental constituents were emitted within irregular shaped euhedral fly ash and were classified as "Group Ca-, Si-,S-,N-, and Fe-rich" in the smoke particles. Ca, Cl, S, and Mg were more abundant in pine smoke than debris smoke. During smoke evolution the evaporated inorganic compounds containing K, Cl, and S condense into potassium salts associated with the "Group K, Cl-rich". This prominent feature distinguishes flaming from smoldering combustion, thus indicating early stage low-temperature condensation processes during aerosol formation. Mixed combustion yields both soot agglomerates and organic particles in C and O-containing groups, reproducing a case of natural fires with initial flaming followed by smoldering.

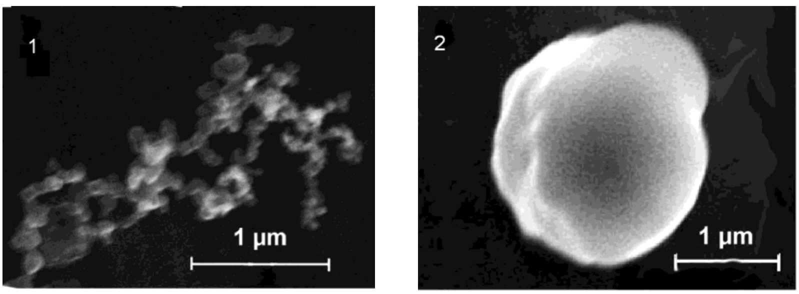

Fig.1. SEM micrographs of typical particles in Group Soot 1) from debris flaming and Group Organic 2) from debris smoldering burns.

4. Carbon fractions and ions

EC and OC contents produced under smoldering, flaming, and mixed conditions are shown in Fig.2. Independent of biomass type, emissions from smoldering burns were characterized by very high OC/EC ratios in the range from 62-194, substantially higher than the ratios observed in the flaming phase, where OC/EC ratios were found in the range from 0.32-0.57. Carbon fraction analysis supports the identification of particle types representative of the burning phase and biomass type. EC particles with 100% C content compose around 25% of Group Soot from flaming fires, in good correlation with the high EC fraction in total carbon (TC) content and low OC/EC ratios. Smoldering burns of pine and debris produced exclusively organic-containing particles with extremely high OC/EC ratios. Measurements of carbon fractions from mixed combustion showed ratios between the ones obtained for smoldering and flaming phases. EC composed around 0.4% of PM mass in smoldering and 10% in flaming burns for both biomass types, consistent with several studies, reporting that flaming fires emit more absorbing BC-rich smoke than smoldering burns (Hopkins et al., 2007).

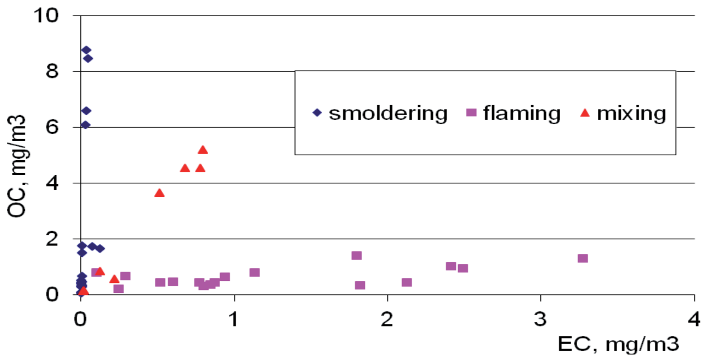

Fig. 2. OC and EC from smoldering, flaming and mixed burns of pine wood and debris.

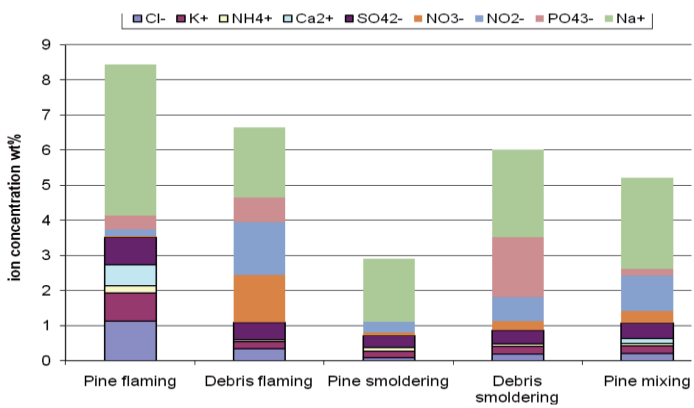

Fig. 3. Ion weight percentages in PM2.5 from various combustion phases.

Inorganic ions in pine and debris-burning particles produced in various combustion phases are shown in Fig. 3. Calcium and potassium combined with sulfates, chlorides, phosphates, and nitrates composed inorganic salts found dominantly in flaming burns, while low temperature combustion during smoldering burns was not apparently sufficient for salt formation. During smoke evolution, uncombusted volatile organics were likely responsible for an increase in the OC fraction of Group Soot, while inorganic compounds were condensed preferably as KCl and K_2SO_4 particles. This phenomenon was observed exclusively for flaming burns; therefore, Group K, Cl-rich may serve as a micromarker of transformation of high-temperature wildfire emissions.

5. Organic/inorganic composition

FTIR spectroscopy complements OC and EC characterization and molecular-level measurements by providing information about functional groups and classes of compounds for the entire aerosol chemical composition. Examination of surface chemistry of fire-produced aerosols showed significant influence of combustion conditions. Under low the temperatures encountered during smoldering fires, the formation of acid and non-acid carbonyl groups in carboxyl acids, ketones, aldehydes, esters, lactones and anhydrides, of hydroxyl groups in anhydrosugars, carboxyl acids, alcohols and phenols, and of aliphatic group–containing compounds were dominant. In contrast, at the high temperatures of flaming burns the polyaromatic and nitro-compound groups were more significant.

Fires conducted in the LAC were characterized by high levoglucosan concentrations (up to 25% of PM mass) from pine wood smoldering burns. Low anhydrosugar emissions during debris smoldering and nearly absent from pine flaming burns confirm levoglucosan as a good molecular marker for wood smoldering. The ratio of Levoglucosan/Mannosan (~ 2-4) was found as an indicator of softwood burning in particular, in accordance with previous studies (e.g., Engling et al., 2006). Other commonly used softwood burning markers, such methoxy phenolic and dehydroabietic acids, were found at lower concentrations and can be used as additional specific tracers for softwood burning.

6. Conclusions

The LAC experiments reported here fill gaps in available data on particulate emissions from Siberian boreal forest fires. Combustion phase was shown to affect the emissions and physico-chemical properties of smoke particles. The high temperatures encountered in open flaming fires significantly influenced the particle formation, producing soot and fly ash. Formation of quasi-liquid tar, organic and fly ash particles at low temperatures was characteristic for smoldering burns. The presented comprehensive approach leads firstly to the quantification of key aerosol parameters and the identification of smoke microstructure through morphological and chemical micromarkers. It also enables multicomponent Siberian biomass burning aerosols to be classified with respect to emission source. This information may be further utilized, through incorporation into source apportionment and global climate models, for estimations of wildfire emission impacts on air quality and climate.

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