# Micromarkers of source-specific combustion aerosols

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Aerosols generated by incomplete combustion of fossil fuels (FF) and biomass burning (BB) are major air pollutants. Open BB relating to wildfires contributes to almost a half of the combustion aerosol global inventory. Aerosol emissions of transport systems are currently acknowledged to be a largest source of uncertainties in understanding the impacts on the regional air quality. Ship emission is among the world's highest polluting sources per quality of fuel consumed. Combustion aerosols may serve tracers allowing a link to distinct combustion sources in apportionment studies if their key physico-chemical characteristics representative the emission sources are defined. The estimations of the source contribution to ambient particulates assume specific markers different from other combustion sources.

A complexity of multicomponent and source-specific PM emissions requires the characterization of individual particles. The classification of combustion aerosols may be done by representative particle types according to their chemical specification. This approach allows quantifying the various diesel and biomass burning sources and identifying the specific characteristic features that can serve as micromarkers emission source.

Presently, the hierarchical cluster analysis (HCA) is a data analysis technique which is used for the interpretation of electron microprobe analysis data and the determination of particle types in ambient air with a large number of pollution sources. Here, we present the development of HCA for characterization the FF and BB emissions by classification of individual particle data on characteristic groups with similar morphological and chemical composition according to the source and type of fuel used. Scanning electron microscopy (SEM) coupled with energy-dispersive X-ray spectroscopy (EDX) is used for characterization of morphology and composition of a large number of individual particles. FTIR spectroscopy and ion content measurements provide the bulk composition data for clustering expert analysis that assures that the mean composition of a cluster is close to physico-chemical identifiable type of particles. Impact of Fe content in laboratory-produced soot aerosol on its composition and structure was analysed in Bladt *et al.* (2012) to demonstrate the elaborated approach for separation individual particles into characteristic groups representing the particle types in dependence of iron content in soot.

This review presents micromarkers of source-specific combustion aerosols emitted by transport systems (road and marine diesel engines) and in open fire experiments. Microscopic and chemical characterization of Opel Astra diesel engine exhaust operated at 35 km/h speed and used diesel B0 fuel with various lube oil (low and high SAPS) and biodiesel fuel B20 with high SAPS are performed. The most abundant group (around 50%) in each exhaust is polluted soot. In diesel B0 exhaust particles, low SAPS groups of silicates, iron and aluminum oxides are found. Using high SAPS with larger ash content leads to abundance of groups of Al oxide and Al fused with Fe. The most prominent impact of high SAPS using relates with presence of well-separated groups of Nb oxides and W particles. Biodiesel exhaust particles, high SAPS contain groups of Fe and Zn oxides, and W particles. The source specific micromarker of biodiesel particles is a separate group enriched by Cl, K, and Na, the elements which are commonly recognized as biomarkers of biofuel combustion.

Exhausts from the main and auxiliary engines operated onboard of a RoRo/casetter carrier ship during TRANSFORM campaign, burning heavy and distillated fuels, respectively, are analyzed (Popovicheva *et al*., 2012). SEM/EDX observations of emitted aerosols indicate carbonaceous particles internally and externally mixed with inorganic species in the combination of spherical and irregular shapes. The composition of individual particles is varying in a wide concentration range of major elements (C, O) and impurities (S, Fe, Si, Ca, Zn, V, Ni, K, Mn) representative the origin of fuels. In distillated fuel (DF)-derived aerosols we identify groups of soot, Ca- and Fe/Si-rich particles. Calcium sulfate/oxide/carbonates and iron oxides are found to compose the group of Ca- and Fe-rich particles, respectively, while silicon oxide and silicate determine the group of Si-rich particles in heavy fuel (HF)-derived emission. Transition metals (V, Ni) containing particles specify the separate group which may serve as a specific micromarker of heavy fuel combustion.

Characterization of freshly-generated smoke particles produced in small-scale spruce open fire experiments shows the groups of soot, Ca- and Fe-rich particles while K- and Mn- rich groups identify the specific micromarkers of BB.

Thus, state-of-the-art characterization of complex multicomponent aerosols improves the quantification of contribution of various fossil fuel and biomass burning emissions to ambient particulates and the identification of a source type in the apportionment studies.

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