# Long-Term Observations of the Sub-micron Aerosol Chemical Composition in the Boreal Forest Reveal a Distinct Bimodal Seasonal Cycle

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Keywords: on-line measurements, aerosol chemical composition, organic aerosol, source apportionment
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## Introduction

The Station Measuring Ecosystem-Atmosphere Relations (SMEAR-II, Hari & Kulmala, 2005) located in Hyytiälä, Southern Finland, is well known among atmospheric scientists due to the large variety of continuous measurements at the station aiming to quantify fluxes, storages and concentrations in the land ecosystem–atmosphere continuum. Aerosol size distribution measurements started in 1996 at SMEAR-II and since then numerous continuous measurements have accompanied this instrument to collect information about aerosol loading, precursors and properties. The long-term measurements of aerosol non-refractory (NR) chemical composition started in early 2012. Before this, a long-term analysis of aerosol composition at the station was only indirectly derivable from aerosol precursor loadings, and aerosol properties such as hygroscopicity and extinction. However, direct measurements are obviously more reliable and necessary to quantitatively explain aerosol sources as well as their seasonal cycles and future trends. This information is needed to diminish uncertainties attributed to aerosol-induced effects on the Earth's radiative balance. This study presents a comprehensive analysis of the seasonal variation of different NR aerosol chemical species in the sub-micron particulate matter (PM) collected at the SMEAR-II. Here we finally provide the missing description of fundamental aerosol characteristics at the station by displaying seven years of on-line chemical composition data and its seasonal variation.

## Methods

The NR PM chemical composition was measured utilizing an Aerosol Chemical Speciation Monitor, ACSM, (Ng *et al.*, 2011). This instrument provides quantitative information, with only a 30 min time resolution, of NR chemical species' loadings: i.e. the concentrations of organics, $NO_3$, $SO_4$, $NH_4$ and and Chl in the ambient sub-micron particles. In the current study we present seven years of data. The aerosol source apportionment was performed utilizing ZeFir pollution tracker (Petit *et al.*, 2017) for direction analysis and Positive Matrix Factorization with the Source Finder toolkit (SoFi, Canonaco *et al*., 2013) for organic aerosol.

## Conclusions

We observe a bimodal trend in the NR PM loading: the largest mass concentrations are detected during summer and winter, the summertime peak being the most significant one. This summertime concentration maximum is linked to enhanced emissions of biogenic volatile organic compounds yielding secondary organic aerosol (OA) upon oxidation. Indeed, during summer months OA makes up nearly 80% of the NR PM mass concentration. In the exceptionally hot Julys in 2014 and 2018 (2.55 and 3.58°C above the 7-year mean, respectively) the organic mass loading was 1.5 and 1.75 times greater compared to the 7-year July OA mean, respectively. Long-term measurements are essential to better understand the effect of heat waves and other abiotic or biotic stress factors on the biogenic OA loading.

The wintertime peak, sharply peaking in February, is greatly influenced by inorganic aerosol species as sulfate forms up to 35% of the total NR PM loading while the organic contribution decreases to ~45%. Overall, the wintertime aerosol is mainly long-range transport and the organic mass spectrum indicates a clear anthropogenic influence. The increased inorganic loading can be linked to a shallower atmospheric mixing layer into which the aerosol precursors are emitted, as well as to the increased need of domestic heating. Future will show how the upcoming push towards renewable energy forms (e.g. carbon neutral Helsinki 2035) will influence the wintertime PM loading.

This work was supported by European Research Council (Grant 638703-COALA).

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