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Online CHARON PTR-ToF-MS measurements elucidate residential heating as the major contributor of wintertime organic aerosol in Fairbanks, Alaska

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Fairbanks, central Alaska, is a sub-Arctic city that frequently suffers from non-attainment of national air quality standards in the wintertime due to the coincidence of weak atmospheric dispersion and increased local emissions but large uncertainties exist about aerosol sources and formation. We determined the general composition and mass concentrations of atmospheric sub-micron non-refractory particulate matter (NR-PM₁) and quantified the sources involved in its formation during a 6-week campaign in the urban centre of Fairbanks in January-February 2022. As part of the Alaskan Layered Pollution and Chemical Analysis campaign (ALPACA), we deployed a Chemical Analysis of Aerosol Online (CHARON) inlet coupled with a proton transfer reaction - time of flight mass spectrometer (PTR-ToF 6000 Ionicon Analytik) and high-resolution aerosol mass spectrometer (HR-ToF-AMS Aerodyne) to measure organic aerosol (OA) and NR-PM₁, respectively, with high temporal resolution of ≤ 1 min. We performed positive matrix factorisation (PMF) of the measurements to apportion aerosol mass to potential sources. The ability of HR-ToF-AMS to measure inorganic species (ammonium, nitrates, sulphates, and chlorides) creates an opportunity to gain insights into their mixing with OA and chemical dynamics. Campaign-averages of OA measured with the two instruments agreed reasonably well ($R^2 = 0.60$) with a regression slope of 0.46. Higher OA concentrations observed with the HR-ToF-AMS are attributable to the particle size-dependence of the CHARON inlet, since it is more sensitive to particles >150 nm; a regression slope approaching 1.0 was achieved for larger primary OA emissions from biomass burning. On the one hand, in line with known emission patterns in Fairbanks, PMF indicated residential heating (Res-H) to be the single largest source of OA here during the wintertime as per CHARON PTR-ToF-MS measurements. A large fraction of OA originated from the combustion of different types of wood ($2.07 \pm 2.47 \mu\text{g}/\text{m}^3$; $47.15 \pm 20.15\%$) and fuel oil ($0.64 \pm 0.64 \mu\text{g}/\text{m}^3$; $16.20 \pm 9.73\%$) as determined from molecular composition and temporal correlation with external tracers (e.g., trace gases and marker ions). On the other hand, using HR-ToF-AMS data, neither OA, nor the overall NR-

PM₁, could be delineated into multiple Res-H sources, and only a single Biomass Burning (BB)-OA related factor could be identified. With the HR-ToF-AMS data, hydrocarbon-like OA (HOA) also appeared as another important contributor to OA ($2.08 \pm 2.79 \mu\text{g}/\text{m}^3$; $38.07 \pm 20.38\%$), other than BBOA ($1.47 \pm 1.81 \mu\text{g}/\text{m}^3$; $28.01 \pm 18.91\%$). In addition, while HOA (or traffic), cooking, and BBOA emissions are almost completely (> 90% of mass) composed of organics, the PMF analysis revealed inorganic aerosol to be distributed across two secondary aerosol factors, i.e., sulphur-rich oxygenated OA and ammonium nitrate, as well as an acidic sulphate aerosol factor. The insights obtained here demonstrate that CHARON PTR-ToF-MS not only provides robust quantitative information but, when combined with a suitable complementary instrument, it generates more refined evidence-based understanding of the dominant sources of OA and processes forming NR-PM₁ which are key to endorsing policy and citizen efforts for the prevention and control of air pollution in Fairbanks, and in the wider Arctic winter.