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Relating refractory soot mass spectra with nanostructure and combustion conditions

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The soot particle aerosol mass spectrometer (SP-AMS) can detect infrared-light absorbing refractory aerosol particles. Vaporization occurs when a focused beam of particles enters a cavity ring down laser (Nd:YAG 1064nm) and the two beams overlap. Refractory mass spectra of soot have previously been used mostly to characterise the ratio of organic to refractory black carbon in soot particles. However, recent studies highlight the information contained in the carbon cluster (Cᵢ⁺) distribution of soot particle refractory mass spectra (Malmborg et al., 2017; Onasch et al., 2015).

Soot mass spectra show strikingly different Cᵢ⁺ distributions depending on the source and combustion condition. Soot in fuel-rich flames at high temperatures and with well-organized nanostructures, typically traditional diesel combustion soot, have mass spectra completely dominated by a few carbon cluster ions with low carbon number (C₁⁺-C₅⁺) of which C₃⁺ has the strongest signal. On the other hand, soot with less organized nanostructure, for example soot from advanced diesel combustion, show a wider distribution of carbon clusters. Soot from these latter types of combustion processes have elevated signals at C₁₀⁺-C₁₅⁺ and C₃₀⁺-C₅₀⁺. The highest signal intensities from carbon clusters with large carbon numbers (i.e., C₃₀⁺-C₅₀⁺) are found at carbon numbers of stable fullerenes (e.g., C₁₅₁₄, C₃₅₀, C₆₀₆, C₇₀₇).

Refractory mass spectra of soot produced in a large number of different combustion processes were collected with the SP-AMS (Aerodyne Research Inc.). The soot nanostructure was analysed using high-resolution transmission electron microscopy (HR-TEM) for a selection of the soot samples. A thermodenuder (Aerodyne Research Inc.) was used, for a subset of the soot samples, to evaporate semi-volatile components.

The C₃⁺ distribution of mini-CAST (Jing Ltd.) OP1 soot with well-organized nanostructures and long lamellae was confined to low carbon numbers. Such distributions were also present for soot from conventional diesel and biodiesel (rape seed methyl esters) combustion in a heavy duty diesel engine without exhaust gas recirculation (EGR). With increased nitrogen flow and decreased oxidation airflow, the C₃⁺ distribution of mini-CAST OP5, OP6 and OP7 soot had increasing signal from large carbon clusters. The mini-CAST soot at OP5-OP7 have decreasing degree of ordered nanostructures and much shorter lamellae than OP1 soot. Similar types of Cᵢ⁺ distributions were found in low temperature diesel combustion at very high EGR (Figure 1).

The spectrum of soot from combustion (i.e., diesel vs biodiesel) influenced the soot nanostructure. However, the C₃⁺ distribution of soot mass spectra from the two fuels was similar, although combustion with conventional diesel had higher fullerene signal. This could indicate that combustion temperatures (which are reduced by EGR) are more important in determining the soot properties that will form large carbon clusters, than fuel composition.

A first analysis between the tortuosity and fullerene signal did not show a correlation. However, it should be noted that short lamellae and highly disordered nanostructure may influence the determination of the tortuosity. Our analysis shows that a higher degree of disorder and shorter lamellae are associated with larger carbon clusters and a wider C₃⁺ distribution. With the large set of refractory mass spectra collected from different soot, our aim is to determine the relationship between soot nanostructure and refractory mass spectra, and the combustion conditions that form the various soot properties.

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