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Ageing Processes of Vehicle Exhaust Aerosols Investigated Through Coupled Aerosol Mass Spectrometer-Thermal Denuder Chamber Measurements

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The aim of this study was to investigate the ageing processes affecting vehicle exhaust aerosols. Since the atmospheric aerosol generally contains a large organic fraction, it is important to consider the properties of organic aerosols (OA) when investigating the impact of particulate matter on climate and human health. Volatility is a key property for understanding OA, as it will determine the atmospheric fate of emissions through influencing gas-particle partitioning (Huffman et al 2009). A major objective of this campaign was the atmospheric relevance of the conditions of ageing processes investigated, the goal being capturing the mechanisms affecting the exhausts after emission.

Vehicle exhaust was selected as it constitutes a significant contribution to ambient OA. For this purpose exhaust was injected into a 6m³ Fluorinated Ethylene Propylene (FEP) bag and irradiated by UV lights for 5h. In most experiments an (NH₄)₂SO₄ seed aerosol was used to reduce wall losses of condensing vapours. Furthermore, exhaust aerosol emitted into the atmosphere will generally mix with ambient particulate mass (PM). The time evolution of the aerosol particle properties of chemical composition, size and mass distributions and hygroscopicity were measured, as well as that of the gas phase chemistry. For a description of the total instrumentation of these experiments, see Nordin et al 2010.

On the timescale permitted by experimental conditions (approximately 6h, the main limitation being the volume of the FEP bag) significant changes in particle chemical composition and volatility were observed through High Resolution Time of Flight Aerosol Mass Spectrometer (HR-ToF AMS) measurements coupled with a thermal denuder. The general trend was decreased volatility and increased oxygen content with time.

| Time | 1h | 2h | 3h |
|-----------|------|------|------|
| RMF 100 C | 0.38 | 0.51 | 0.56 |
| RMF 70 C | 0.65 | 0.85 | 0.90 |

Table 1. Remaining Mass Fraction (RMF) of particulate organic compounds after 1, 2 and 3h of UV irradiation.

Formation of secondary PM through condensation was observed, which implies reduced volatility of emitted gas phase species, possibly due to oxidation.

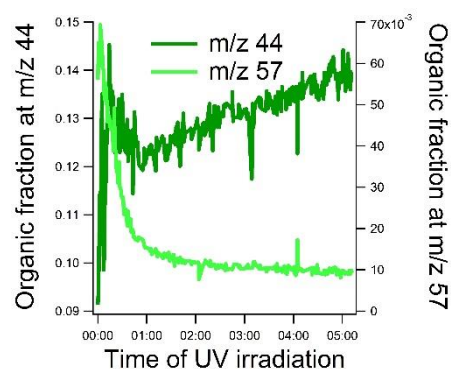


Figure 1. Exhaust aerosol ageing as parameterized by AMS organic signal fraction at m/z 44 and 57.

The figure and table above shows that exhaust aerosol produced by a Volvo V40 (1998) ages significantly on a timescale of hours. The signal at m/z 57 is mainly due to C₄H₇⁺, a fragment often considered a tracer for fresh engine exhaust. The immediate decline of this fraction under irradiation highlights the need for highly time-resolved measurements. CO₂⁺ causes the organic signal at m/z 44, this fragment is produced by a vast number of organic compounds and used here as a crude marker for increased oxygen content. The increase in m/z 44 implies that further development occurs beyond the time captured by this experiment. Considering the observed gas to particle transformation, and given the fact that reactions generally occur faster in gas phase, the chemical properties of condensing emissions likely account for these trends.

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