# Characterization of natural gas engine emission by aerosol mass spectrometer

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The usage of natural gas (NG) engines is expected to increase in coming years due to the increased availability, competitive cost and lower $CO_2$ emissions compared to conventional liquid fossil fuels. However, NG engines can produce emissions that can have impact on environment and human health. The major hydrocarbon species emitted by NG engines is methane and another important emission component found in NG engines emissions is formaldehyde. Particle emissions from NG engines are known to be low compared to conventional diesel engines, because of lower soot particle formation in combustion, but particle number emissions of NG engines, especially nanoparticle emissions, are not necessarily low. In order to diminish the environmental and health effects with the tightening emission limitations the after-treatment systems are used increasingly also with the NG engines. The aim of this study was to examine three different catalyst systems (oxidation catalysts and Selective Catalytic Reduction, SCR) in the natural gas engine operating at different conditions. The effects of catalyst systems on the gaseous and particle emissions were investigated with the special focus on the chemical composition of particles.

Two measurement campaigns were conducted in 2014–2015. The test engine was a passenger car gasoline engine modified to run with NG. The driving conditions were selected based on the emission levels and two different engine driving modes were used. Two catalyst setups were tested the first one consisting of a combination of an oxidation catalyst and a SCR and the other setup having of only one oxidation reactor. Exhaust gas temperature was varied from 350 to 500 °C and exhaust gas flow was either 80 kg/h or 40 kg/h.

The chemical composition of NG emission particles was studied by using a Soot Particle Aerosol Mass Spectrometer (SP-AMS, Aerodyne Research Inc, Onasch et al., 2012). In addition to the particulate chemistry, inorganic and organic gases, particulate matter (PM) and particle size distributions were measured by several instruments. The volatility of particles was investigated by using a thermodenuder and the potential of NG emission to produce secondary particles was examined by Potential Aerosol Mass (PAM)-chamber (Kang et al., 2007). NG exhaust was diluted by a factor of ~10–150 depending on the measurement devices.

The particle measurements indicated that the catalysts decreased the total PM at all test conditions by 45-73%. However, the PM concentrations increased as the exhaust temperature increased. Based on the concentrations of chemical species, the increase of mass was mostly due to the larger concentration of sulfate and ammonium but also hydrocarbon concentration was larger with higher exhaust temperature. Detailed study of the mass spectra of organics revealed that in some cases a large portion of hydrocarbon signal was found at very small mass fragments (e.g. $CH_2^+$, $CH_3^+$, $CH_4^+$) compared to what is typically found in e.g. diesel fuel exhaust particles. In addition to organic and inorganic species (sulfate, nitrate, ammonium), the SP-AMS enabled the detection of metals in NG emission particles.

Regarding the potential of NG emissions to form secondary aerosol, the concentration of potential secondary organic aerosol was more than ten times larger than the concentration of primary organic aerosol in some engine conditions. The ratio of potential secondary aerosol to primary aerosol was even larger for inorganic nitrate, sulfate and ammonium.

The disadvantage of the usage of the SP-AMS in NG emission measurements is the particle size range measured by the SP-AMS. SP-AMS can detect particles from ~50 to 800 nm whereas the number size distribution of NG emission particles can peak at as small particle size as 2-5 nm (Alanen et al.2015). The formation of secondary particles increases the size, which can distort the comparison of primary and secondary particles as the secondary particles as more readily measured with the SP-AMS.

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