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Dilution and aerosol dynamics within a diesel car exhaust plume—CFD simulations of on-road measurement conditions

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Abstract

Vehicle particle emissions are studied extensively because of their health effects, contribution to ambient PM levels and possible impact on climate. The aim of this work was to obtain a better understanding of secondary particle formation and growth in a diluting vehicle exhaust plume using 3-d information of simulations together with measurements. Detailed coupled computational fluid dynamics (CFD) and aerosol dynamics simulations have been conducted for H_2SO_4–H_2O and soot particles based on measurements within a vehicle exhaust plume under real conditions on public roads.

Turbulent diffusion of soot and nucleation particles is responsible for the measured decrease of number concentrations within the diesel car exhaust plume and decreases coagulation rates. Particle size distribution measurements at 0.45 and 0.9 m distance to the tailpipe indicate a consistent soot mode (particle diameter $D_p\sim50$ nm) at variable operating conditions. Soot mode number concentrations reached up to 10^{13} m^{-3} depending on operating conditions and mixing.

For nucleation particles the simulations showed a strong sensitivity to the spatial dilution pattern, related cooling and exhaust $H_2SO_{4(g)}$. The highest simulated nucleation rates were about 0.05–0.1 m from the axis of the plume. The simulated particle number concentration pattern is in approximate accordance with measured concentrations, along the jet centreline and 0.45 and 0.9 m from the tailpipe. Although the test car was run with ultralow sulphur fuel, high nucleation particle ($D_p\leqslant15$ nm) concentrations ($>10^{13}$ m^{-3}) were measured under driving conditions of strong acceleration or the combination of high vehicle speed (>140 km h^{-1}) and high engine rotational speed (>3800 revolutions per minute (rpm)).

Strong mixing and cooling caused rapid nucleation immediately behind the tailpipe, so that the highest particle number concentrations were recorded at a distance, $x=0.45$ m behind the tailpipe. The simulated growth of H_2SO_4–H_2O nucleation particles was unrealistically low compared with measurements. The possible role of low and semi-volatile organic components on the growth processes is discussed. Simulations for simplified H_2SO_4–H_2O–octane–gasoil aerosol resulted in sufficient growth of nucleation particles.

Keywords: Aerosol dynamics modelling; Particle formation; Particle growth; Soot; Vehicle exhaust plume

1. Introduction

Adverse health effects such as lung cancer (e.g. Pope, 2000), cardiovascular disease (e.g. Peters

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et al., 2001) bronchitis, asthma (Peters et al., 1997) and mutations (Somers et al., 2004) are associated with respirable particles emitted from vehicles. Ultrafine particles (defined as $D_p \leqslant 100\,\text{nm}$) can deposit with high efficiency in the smallest vessels of the lungs. One major source of these ultrafine particles is vehicles. Furthermore, the influence of traffic-generated combustion particles on the earth radiation budget is controversially discussed (e.g. Jacobson, 2002; Penner et al., 2003).

According to health effect studies, the particle number concentration to which the individual is exposed is more important than their mass (e.g. Donaldson et al., 1998; Sydbom et al., 2001). The deposition efficiency of particles in the smallest vessels in the lung increases with decreasing particle size. For this reason and the fact that ultrafine particles contribute much to number and little to mass emissions, it is important to study the formation and fate of ultrafine particles in the exhaust plume of vehicles. Thereby, various processes such as dispersion, coagulation, deposition, new particle formation and growth must be assessed.

Diesel engines primarily emit aerosol precursor gases such as SO_2, SO_3, H_2SO_4, NO_x, H_2O, low and semi-volatile organic species as well as soot particles. At the engine exhaust port, soot particles are fractal-like agglomerates, which undergo complex processes like compaction and condensation of low and semi-volatile organics, which result in higher density and less irregular shape until they leave the exhaust pipe. Exhaust size distribution measurements made at various diesel cars are similar for comparable operation conditions unless nucleation occurs. They exhibit a consistent soot mode at a particle mobility diameter of 50–100 nm (e.g. Harris and Maricq, 2001). Dependent on fuel composition, engine operating conditions, exhaust aftertreatment, exhaust dilution and sampling methods, the size distribution can exhibit a second highly variable nucleation mode with diameters from 10 to 20 nm (e.g. Maricq et al., 2002; Vogt et al., 2003; Giechaskiel et al., 2005). Earlier works all undertaken on heavy-duty diesel engines resulted in qualitatively similar results (Baumgard and Johnson, 1996; Khalek et al., 1999; Shi and Harrison, 1999). Nucleation particles can dominate the number concentration of ultrafine particles. In contrast to soot mode particles, there is only limited information available on the chemical composition of nucleation mode particles. Volatility tandem DMA (VTDMA) measurements suggest that most of these particles are semi-volatile and do not have a solid core (Scheer et al., 2005; Maricq et al., 2002). Highway roadside measurements by Wehner et al. (2004) showed a bi-modal distribution with a volatile nucleation mode and a small volatile fraction of soot mode particles. The nucleation mode is thought to originate from sulphuric acid and/or semi-volatile organic gases that nucleate and condense during exhaust dilution. There is strong evidence that nucleation particles in vehicle exhaust may be formed by binary homogenous nucleation of sulphuric acid and water. In several studies, high particle number concentrations were related to high fuel sulphur content and exhaust aftertreatment such as the use of oxidation catalysts (e.g. Baumgard and Johnson, 1996; Maricq et al., 2002; Vogt et al., 2003). Oxidation catalysts remove a large fraction of hydrocarbons and boost SO_2 to SO_3 oxidation. SO_3 then rapidly reacts with exhaust H_2O forming sulphuric acid. Further suggested particle formation mechanisms are homogeneous ternary nucleation of H_2SO_4, NH_3 and H_2O (Shi and Harrison, 1999) and heterogeneous ion-induced nucleation (Yu, 2001). However, the relevance of these mechanisms is still not clear. Organic compounds are considered to control the growth of nucleation particles in diesel exhaust plumes (Khalek et al., 2000; Tobias et al., 2001; Sakurai et al., 2003).

A key factor for nucleation is the dilution of exhaust gases. During dilution, aerosol precursor gases cool and the saturation ratios of semi-volatile species go through a maximum, which may lead to nucleation and/or condensation. Engine test-bed investigations indicate that the occurrence of nucleation particles strongly depends on dilution ratio, residence time, dilution temperature and relative humidity. However, Maricq et al. (2002) and Vogt et al. (2003) encountered difficulties when comparing laboratory data to either wind tunnel or on-road (e.g. chasing) measurements. Giechaskiel et al. (2005) concluded that vehicle exhaust nucleation mode particles can be at least qualitatively reproduced in the lab.

In principle, a coupled CFD-aerosol model approach enables to account for all relevant concurrent processes in the exhaust plume, including turbulent diffusion. Moreover, 3-d pattern of all parameters of interest are calculated and can be used for validation with point measurements and further interpretation due to the 3-d information.