Sulfur driven nucleation mode formation in diesel exhaust under transient driving conditions

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4 Figures

Supporting information

Detailed experimental setups for all the four experiments performed are shown in Figures S1&S2. In the engine laboratories (Fig. S1a) and in the aerosol laboratory (Fig. S1b) the primary exhaust gas dilution was executed with a porous tube diluter (PTD). In the PTD the dry and filtered dilution air flow was 50 l/min, the temperature was 30 °C and the dilution ratio was maintained close to a constant value (12). With these parameters, nucleation mode formation was aimed to maintain repeatable. In the engine laboratories, the primary dilution ratio was determined by measuring the CO_2 concentrations both in hot exhaust and diluted exhaust. In the aerosol laboratory, the SO_2 concentrations were used in the dilution ratio determination. The secondary dilution was performed by an ejector type diluter (Dekati diluter) in order to reach suitable concentration levels for the instruments. The instrumentation consisted of Scanning mobility particle sizers (SMPS, TSI Inc.), both the Nano-SMPS and the SMPS in Fig. 1a and only the Nano-SMPS in Fig. 1b, and an Electrical low pressure impactor (ELPI, Dekati Oy) with extra impactor and filter stages. The instruments enabled particle number size distribution analysis over a broad particle size range (3 nm – 10 μm). Occasionally, a thermodenuder with low nanoparticle losses[1] was applied to test the aerosol upstream of the instruments.

Solely in the engine laboratory study of the heavy-duty diesel engine (experiment 2), the gaseous sulphuric acid (GSA) concentration was measured using a Chemical Ionization Mass Spectrometer (CIMS) setup which consisted of a flow tube reactor and an ion trap mass spectrometer. In the flow tube reactor, gaseous H_2SO_4 molecules undergo an ion-molecule reaction with the reagent ions (NO_3-HNO_3), leading to product ions of the HSO_4-HNO_3 type. The GSA concentration is determined from the abundance ratio of product and reagent ions measured with an ion trap mass spectrometer. Details of the instrument can be found in Speidel et al. (2007)[2].

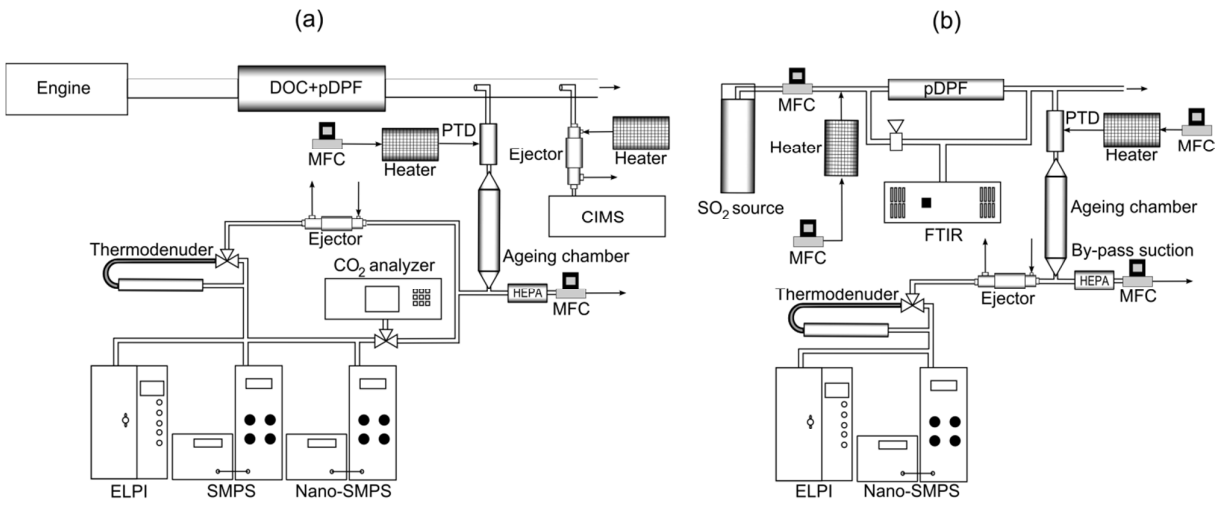

Figure S1. Experimental setups in the engine laboratory tests (a) and in the aerosol laboratory (b). The CIMS and the adjacent ejector diluter were only used in the heavy-duty engine laboratory experiment (experiment 2).

The nucleation mode particle characteristics were studied in the aerosol laboratory (experiment 1). The volatility of nucleation mode particles was tested by measuring the Nano-SMPS size distribution with and without thermodenuder treatment (at 265 °C). The particle mode was completely eliminated by the thermodenuder treatment, as presented in Rönkkö et al. 2011[1]. The particle charging state was studied by applying an efficient electrostatic precipitator with high voltage (3.75 kV) in the inlet of the Nano-SMPS (flow 1.5 l/min). The particle collection efficiency of the EPS was close to 100% for singly charged particles at nucleation mode size range. There was no change in the measured particle concentrations whether the high voltage was on or off (Fig. S2).

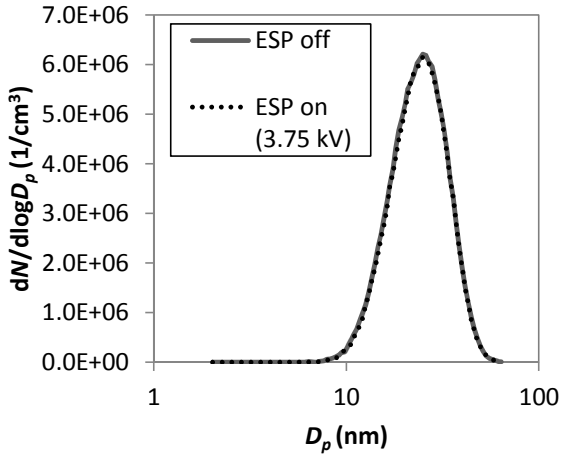

Figure S2. Nucleation mode particle size distributions in experiment 1 measured with and without an electrostatic precipitator (ESP).

Particle concentrations of the exhaust plume were measured by chasing the truck with a trailer by the "Sniffer" mobile laboratory van. Using a laser based distance-meter, the distance between the truck and the laboratory vehicle was kept constant at 12 ± 2 m. The probe for sampling the exhaust plume aerosol was located above the front bumper of the laboratory vehicle. The sample was conveyed continuously to the particle instruments. The particle instrumentation inside the mobile laboratory consisted of an ELPI, an SMPS, a Nano-SMPS and an ultrafine CPC. Furthermore, thermodenuder treatment (at 265°C) was occasionally applied to the aerosol sample. The laboratory van was also equipped with gas analyzers for CO_2, CO and NO_x.

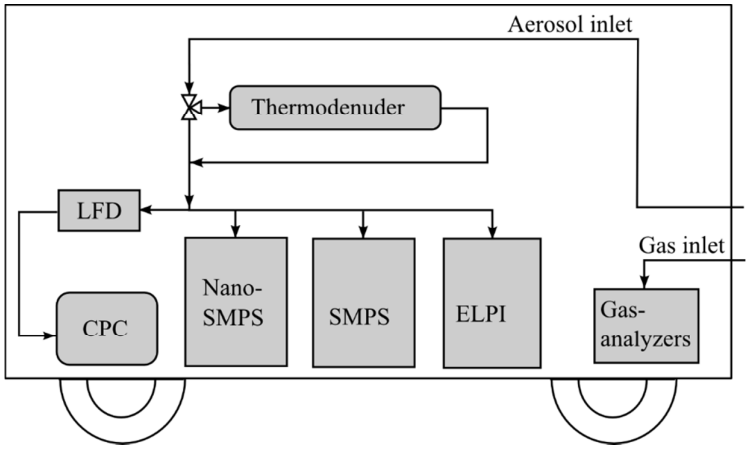

Figure S3. Experimental setup in the mobile laboratory.

Studies were conducted on Highway 4, Finland, between Oulu - Jyväskylä - Oulu. The distance between the two cities is 350 km. The long route was chosen in order to get statistical data and, in order to avoid the effects of other traffic, the measurements were performed during the late evening and at night. Fig. S4 shows the altitude profile for the test route. Near Jyväskylä, the test period consisted mainly of uphill and downhill driving conditions, while near Oulu the route was relatively flat. However, the vehicle speed was kept near 80 km/h throughout the test route. Although speed was relatively constant, engine load varied strongly, depending mainly on the altitude profile; during uphill driving engine torque was typically at maximum and during downhill it was zero, meaning driving at engine braking conditions. Here "torque" refers to the torque produced by the engine. The negative torque caused by the inertia of the truck was not measured.

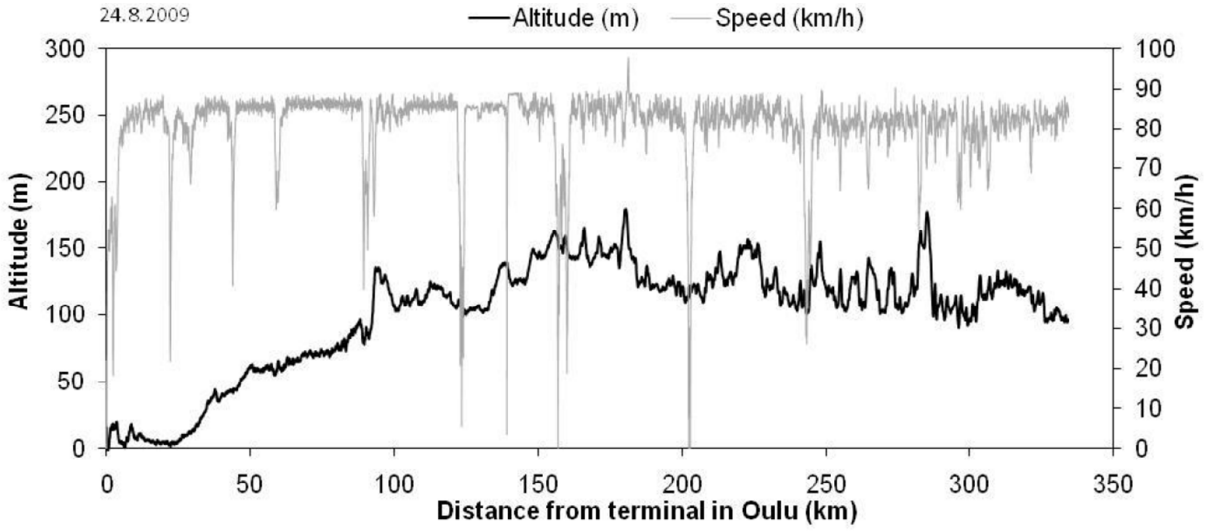

Figure S4. The speed and altitude profiles on the transportation route of the test truck (Oulu – Jyväskylä, Finland).

References

1. Rönkkö, T.; Arffman, A.; Lähde, T.; Karjalainen, P.; Heikkilä, J.; Pirjola, L.; Rothe, D. and Keskinen, J. Diesel exhaust nanoparticle volatility studies by a new thermodenuder with low solid nanoparticle losses, *15th ETH Conference on Combustion Generated Nanoparticles* **2011**.

2. Speidel, M.; Nau, R.; Arnold, F.; Schlager, H. and Stohl, A. (2007) Sulfur dioxide measurements in the lower, middle and upper troposphere: Deployment of an aircraft-based chemical ionization mass spectrometer with permanent in-flight calibration. *Atmospheric Environment* **2007***,* 41, 2427–2437.