Supporting Information
Traffic is a major source of atmospheric nanocluster aerosol, by Rönkkö et al.

On the background CO_2 concentrations of ambient air

The average atmospheric CO_2 level is currently considered to be at ca. 400 ppm. Atmospheric CO_2 concentration, however, has significant spatial and temporal variation depending e.g. of nearby vegetation[46] and, like in this study, of nearby combustion emission sources. The background CO_2 concentrations display temporal variation as well, typically seen as relatively strong diurnal and seasonal cycles.[47] The variation of local background concentrations of atmospheric CO_2 are seen as plateaus in measurements conducted in street canyon (long measurement time) and in on-road experiment (large spatial variation). In addition, in the on-road experiment the motorway driving caused slight mechanical shaking and tremors to the instrument, which introduced random normally distributed noise component to the data, likely by the electronic components of the instrument. As this random component can be negative as well as positive, also values below the background value were returned by the instrument. In the on-road experiment, the highest number (N) of CO_2 observations belonged to the concentration ranges of 390-400 ppm (28.5%, no elevated NCA concentration) and 400-410 ppm (21.5%, slightly elevated NCA concentration), and a minor fraction of observations (3.5%) was below 380 ppm.

The sideway wind on the bridge crossing the Danish Straits (see Fig 2c) enabled us to avoid the effects of nearby CO_2 sources and sinks to the CO_2 measurement and thus to quantify the local background CO_2 concentration and the above-mentioned driving-caused noise. The mean CO_2 concentration during bridge driving was 388.9 ppm (median value was 389.3 ppm), while the standard deviation of the CO_2 concentrations was 12.5 ppm. Importantly, the variation of CO_2 concentration during this local background measurement was random and followed a normal distribution.

The above-mentioned variation in background CO_2 concentrations did not affect the NCA emission factors in the main text because those were determined from slopes of increasing NCA concentration.

Computing the total number emissions of NCA globally

In 2013, road transport produced worldwide 5.5 Gt of CO_2.[48] Using the same conversion factors as earlier (3.14 gCO_2/g_{fuel}), we can now convert the emission factors to total global road transport emissions.

For the 'roadside' minimum NCA emission factor of 2.4×10^{15} kg_{fuel}^{-1}, we get as the total emission $((5.5\times10^{12}$ kg/3.14)/a)$\times2.4\times10^{15}$ kg_{fuel}^{-1} = 4.2×10^{27} a^{-1} from road transport alone. This can be compared to the estimated total anthropogenic particle number emissions[32] that estimates worldwide particle number emissions at 17×10^{27}; from this, we see that the annual NCA emission resulted from road traffic is of considerable size (25%) with respect to total emissions.

Figure captions

Fig. S1. The location of the roadside measurement station, the street canyon measurement station and the weather station on (a) the map of the Helsinki Metropolitan area. The exact location of the measurement station and the road influenced (red) and clean (blue) sectors with respect to the wind direction measured at the weather station are shown in subfigures for the roadside (b) and for the street canyon (c).

Fig. S2. Time series of (a) the number concentrations measured by the particle size magnifier (PSM) and the condensation particle counter (CPC), (b) the relative amount of nanocluster aerosol (NCA) and (c) the wind direction. The red and blue colors represent the road influenced and background sectors, respectively, and black indicate the sector between them.

Fig. S3. The profile of the measurement site in the street canyon environment. The sampling location is marked with red dots.

Fig. S4. The mobile unit (a) shown from the side and (b) on road conducting the long-range experiment through Europe.

Fig. S5. Measured number concentrations for particles larger than 3 nm in diameter (upper panel), total particle number concentrations (middle panel) and CO_2 concentrations (bottom panel) measured during the on-road measurements.

Fig. S6. Measured weather data during the on-road measurement. From top to bottom: the air temperature (T), relative humidity (RH), barometric pressure (p) and altitude are shown.

Fig. S7. Measurement setup in the engine laboratory experiments. The aftertreatment system consisted of a diesel oxidation catalyst (DOC), diesel particulate filter (DPF), and a selective catalytic reduction (SCR) system. The sample was diluted with clean pressurized air (PA) in a two-stage dilution system consisting of a porous tube diluter (PTD) and an ejector diluter. The instruments used in the measurements were an electrical low pressure impactor (ELPI), condensation particle counter (CPC), particle size magnifier (PSM), scanning mobility particle sizer (SMPS), and an engine exhaust particle sizer (EEPS).

Fig. S8. Exhaust particle size distributions measured after the thermal treatment of diluted exhaust sample. Particle size distributions were measured by using the particle size magnifier (PSM) (solid lines) and the scanning mobility particle sizer (SMPS) (dashed lines).

Fig. S9. Nanocluster aerosol (NCA) number concentrations in two different roadside environments, semi-urban (solid line) and street canyon (dashed line). In addition to Fig. 1a and 1b (main text), the data for all wind directions is shown (black). The sectors of road (red) and urban background (blue) are distinguished based on the wind direction. (a) The diurnal variation of the NCA concentration and (b) the particle number size distributions are shown.

Table captions

Table S1. Driving parameters (engine load and torque) and regulated emissions of the heavy duty test engine. Filter smoke number (FSN) indicate the particulate mass emission of the engine.