Source Apportionment and Size Characterisation of Aerosol Particles Measured in a Copenhagen Street Canyon

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1. Introduction

It is recognised that particles in urban air are responsible for serious health effects, i.e. long-term effects like cancer and cardiovascular decease as well as acute effects like allergy or irritation of eyes, nose and throat. Especially, the very small particles – ultrafine particles – are assumed to be important for the adverse health effects of particles. Road traffic is the dominating source to particles in urban air in greater cities. While dynamometer cycles are essential to establish uniform emission standards for regulatory purposes and for testing of new technologies, they do not reflect the real driving conditions and the level of maintenance of the actual vehicle fleet. Further, measurements at tail pipe exit or in emission test facilities show size distributions peaking at higher diameters that in ambient urban air. Thus, there is a need for on-road emission estimates of air pollutants from the actual fleet.

2. Characterisation of particles

In order to characterise the particle pollution emitted directly from traffic, a method has been employed, which is based on the Differential Mobility Analyser, DMA, using the separation of particle size fractions by their mobility determined by movement of charged particles in an electrical field. The DMA can measure with high time resolution, which is necessary for identification of the air pollution from traffic, in order to be able to separate this type of pollution from other types of air pollution.

3. The measurements

The measurements were performed in a street canyon close to central Copenhagen (Jagtvej). A fixed monitoring station of the Danish Air Quality Monitoring Programme has been in operation at this location many years (Kemp and Palmgren, 1999). Data from this station include half-hour measurements of NO_x, CO and other traditional pollutants. A 28 cm Vienna-type Differential Mobility Analyser (Winklmayr et al., 1991) using a re-circulating flow system (Jokinen and Mäkelä, 1997) in connection with a TSI Model 3010 Condensation Particle Counter was used to measure particle numbers in 29 contiguous electrical mobility channels in the size range 6-700 nm. A PC was used to control the scanning process. Corrections for 50% average channel efficiency, and for zero and multiple electron charging, were made (Wiedensohler, 1988). The particle counter efficiency drops steeply for particle diameters below 10 nm; a correction for this is made using the efficiency function given in Mertes et al., 1995. No corrections have been made for the diffusion losses in the air inlet nor in the DMA, but the same inlet system and the same operational parameters were used in all the particle measurements reported here. The scanning time for each spectrum was approx. 3 minutes, and average spectra for every half hour were calculated and used for the analysis in relation to half-hour average concentrations of CO and NO_x. The measurements were carried out continuously during two campaigns in the periods 19 January - 8 March 1999 and 28 January - 6 March 2000.

4. Determination of the sources

Receptor analysis of the measured data was performed, using factor analysis (or principal component analysis) for the identification of the important sources. The factor analysis of the weekly average cycle in 1999 (Wåhlin et al., 2001a) showed that petrol traffic and diesel traffic can be identified as two factors, which are responsible for almost all the variation (95 %) of the concentrations of CO, NO_x, and particles in the size range < 300 nm. A third non-traffic factor (insignificant covariance with CO and NO_x), is responsible for some of the variation of the particle concentrations in the size range > 300 nm. This size range indicates that the non-traffic factor is associated with long-range transport of secondary aerosol particles.

5. The results of the receptor analysis

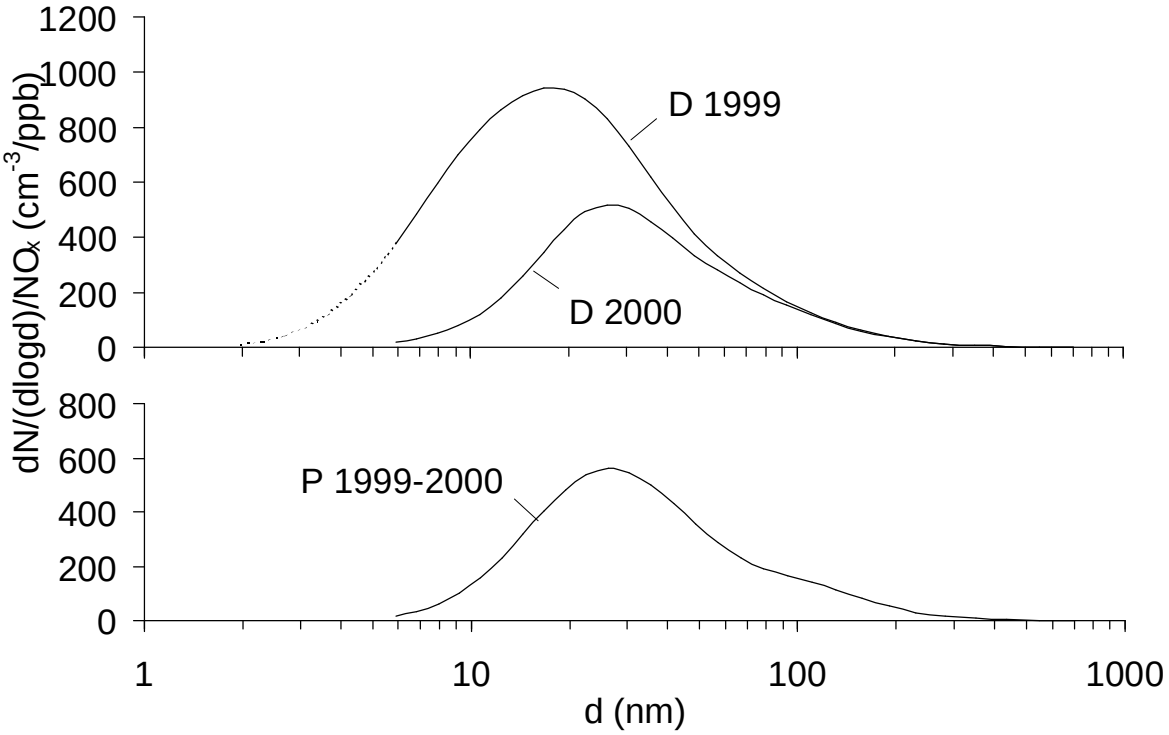

Figure 5.1. The diesel ('D 1999', 'D 2000') and petrol ('P 1999-2000') emission profiles for Jan-Mar 1999 and Jan-Mar 2000.

The high co-variation between particles, CO, and NO_x, and the clear existence of variations in the 1999 data due to differences in diesel and petrol traffic, inspired us to perform a linear "least squares" analysis (multiple regression) of the average week cycle data (Wåhlin et al., 2001b). We used the concentrations of CO and NO_x as the independent variables, while the concentrations of the particles in the different size fractions were considered as the dependent variables. The calculated slopes, with uncertainties due to the scatter, can be used for an assessment of the traffic emission profiles, while the offsets can be considered as non-traffic contributions. Using a CO/NO_x emission ratio of 1 mol/mol as a typical upper limit for diesel traffic (ref. UK Emission Factors Database), it can be shown that the CO emissions from diesel traffic at this monitoring station are insignificant compared with the emissions from petrol traffic. The Particle/NO_x slopes can therefore be considered as good estimates of the average particle/NO_x emission ratios of diesel during the two campaigns. The slopes ('D 1999' and 'D 2000') are plotted in Fig. 5.1. The values of the two campaigns are different in nearly the whole size range < 100 nm, particularly in the nano-size range < 30 nm. We cannot think of any other reason for this phenomenon than the substantial reduction of the sulphur content from 500 ppm to less than 50 ppm in the diesel fuel used in Danish road traffic, which was implemented in July 1999 in the period between the two campaigns. On the other hand, petrol fuel has not changed in this period, and we expect that the petrol traffic emission profiles were similar during the two campaigns. It is remarkable that the positions of the regression planes for the 1999 campaign and the 2000 campaign, although separated, are tilted against each other, so they intersect in a line with a constant CO/NO_x ratio of 33.1±0.6 mol/mol, independent of the particle size. It is natural to consider the intersection line as the average petrol emission profile for both campaigns. The calculated particle/NO_x ratios ('P 1999-2000') for the petrol emission profile are shown in Fig. 5.1. A constrained receptor model, COPREM, was used for the source apportionment, using the technique described in Wåhlin et al., 2000a.

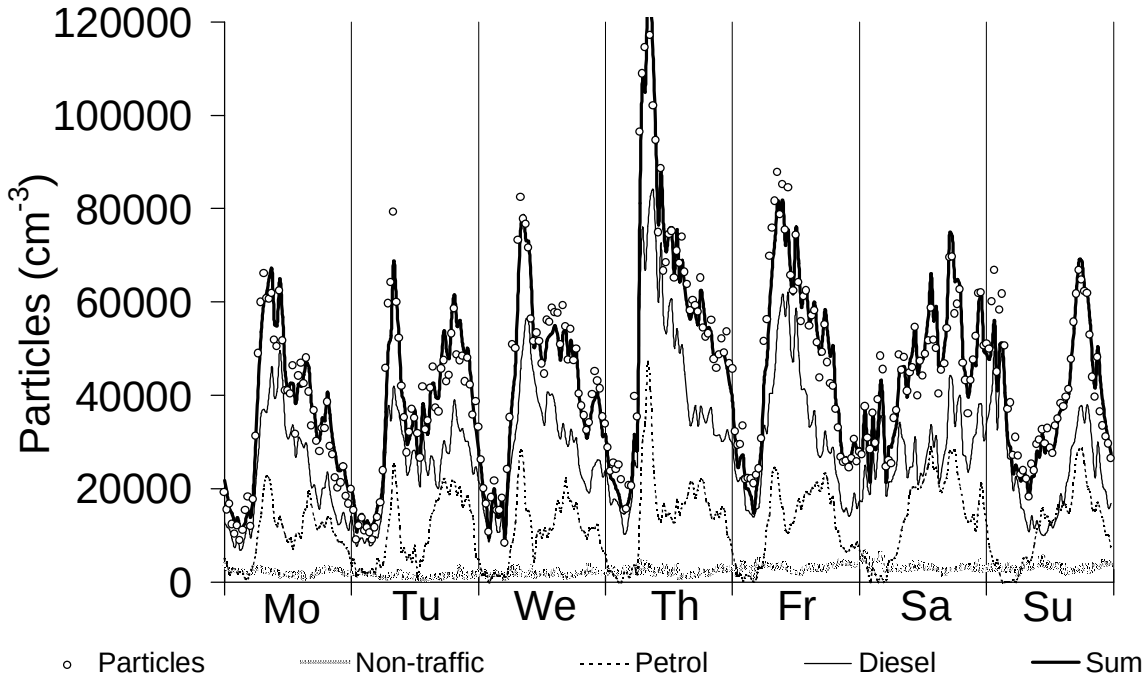

Figure 5.2. The weekly average cycle of particles at Jagtvej in Jan-Mar 1999.

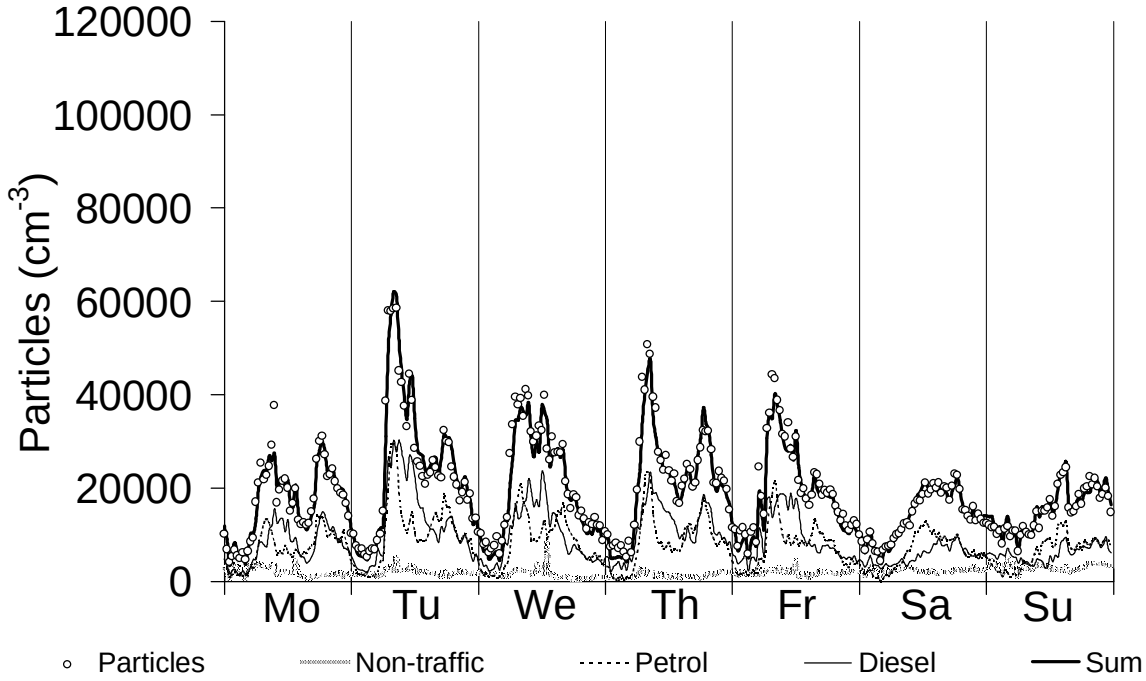

Figure 5.3. The weekly average cycle of particles at Jagtvej in Jan-Mar 2000.

The measured average weekly cycles of total particle concentrations are shown in figures 5.2 and 5.3. Also shown in the figures are the COPREM solutions using the petrol CO/NO_x ratio 33 mol/mol. The average particle size distributions and the COPREM fits during the two measuring periods are shown in figures 5.4 and 5.5.

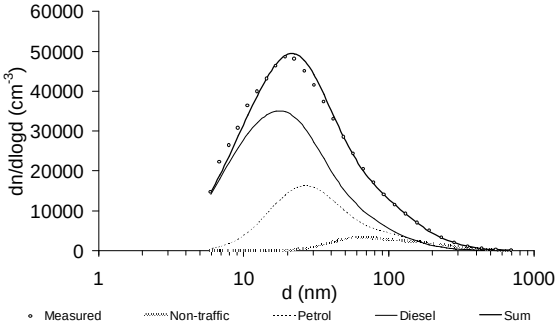

Figure 5.4. The average particle size distribution at Jagtvej in Jan-Mar 1999.

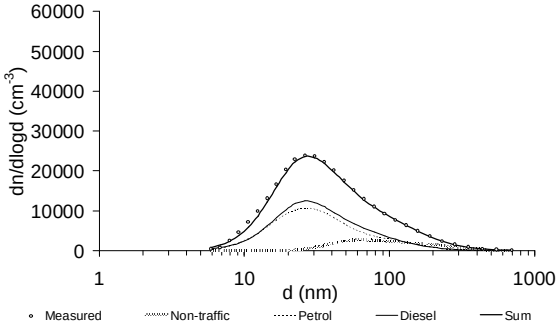

Figure 5.5. The average particle size distribution at Jagtvej in Jan-Mar 2000.

6. References

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