Source apportionment of sub-micron particles in the urban background by Positive Matrix Factorization

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Atmospheric particles are regulated and monitored by a lot of countries as they are strongly associated with poor visibility and adverse health effects. Presently, these regulations are focused on particle mass concentrations. However, other particle metrics, like particle number concentration (PNC) or particle surface concentration, (both dominated by submicron particles) are discussed to be better correlated to some health endpoints than mass concentration, especially for cardiovascular diseases, as was also observed in some epidemiological studies [Atkinson et al., 2010; Breitner et al., 2011]. As a result, there is an ongoing discussion if sub-micron particle metrics should also be subjected to regulation in ambient air [ETH, 2012].

The presented study was started on this background to obtain more detailed source information on particle number concentrations by using particle number size distribution data along with some extra measurements from daily monitoring.

The measurements were conducted at a joined urban background measurement site of IUTA and the State Agency for Nature, Environment and Consumer Protection of North Rhine Westphalia (LANUV) in Mülheim Styrum, Germany. This site is influenced by traffic emission from a motorway in northern directions. The main wind direction is from the southwest. Measurement data for a three-year period from March, 2009 to February, 2012 was selected for further evaluation. Following metrics were determined: number size distribution (14.1 - 736.5 nm), lung-deposited particle surface concentration (LDS), PM_1 (starting from March, 2011), PM_{10} and NOx. Data for PM_{10}, NOx and for meteorological conditions were provided by LANUV.

All data were rigorously evaluated prior to processing. The time resolution of the data was averaged to two-hourly values and the particle size classes were reduced to 16 channels per decade to facilitate the source apportionment analysis. Further, following size classes were used as additional PMF variables: total number concentration (Total), number concentration for ultra fine particles (UFP, <100nm) and number concentration for the particles larger than 100nm (FP).

The receptor model EPA PMF 3.0 was chosen for this study as this software has been successfully applied to similar dataset in a previous study [Gu et al., 2011]. Among all the results from this model, the seven-factor result was considered to be the most robust and plausible based on factor composition, diurnal patterns and conditional probability function (CPF) diagrams of wind direction influence. The identified source groups and their averaged contributions to some source-indicative variables are shown in Table 1.

Table 1 Contributions of resolved factors (%)

Short name	NOx $\mu g\ m^{-3}$	UFP cm^{-3}	FP cm^{-3}	Total cm^{-3}	LDS $\mu m^2 cm^{-3}$	PM_{10} $\mu g\ m^{-3}$
Nucleation	1.3	15.0	0.9	12.9	~0*	0.6
Secondary	19.6	1.6	10.3	2.9	9.1	24.5
Dust	4.9	0.8	3.6	1.2	3.5	16.2
Background	3.1	0.8	31.1	5.2	17.4	32.8
Fresh Gasoline	17.7	36.4	5.4	31.9	16.2	8.2
Aged Gasoline	21.0	33.1	4.7	28.8	21.7	10.1
Diesel	32.4	12.4	43.8	17.1	32.1	7.5

The results show that traffic factors appear to be the major contributors for total and UFP particle number concentrations, while the nucleation factor and the other factors have moderate or low contributions to them. The three distinguished traffic related factors resemble corresponding emission particle size distribution profiles and have typical diurnal patterns. However, without support by chemical analyses, interpretations of factor characteristics may imply considerable uncertainty due to the absence of selective or unique tracer compounds.

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