Advanced source apportionment of size-resolved trace elements at multiple sites in London during winter

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Trace element measurements in PM10-2.5, PM2.5-1.0 and PM1.0-0.3 aerosol were performed with 2 h time resolution at kerbside, urban background and rural sites during the ClearfLo winter 2012 campaign in London. The environment-dependent variability of emissions was characterized using the Multilinear Engine implementation of the positive matrix factorization model, conducted on data sets comprising all three sites but segregated by size. Combining the sites enabled separation of sources with high temporal covariance but significant spatial variability. Separation of sizes improved source resolution by preventing sources occurring in only a single size fraction from having too small a contribution for the model to resolve. Anchor profiles were retrieved internally by analysing data subsets, and these profiles were used in the analyses of the complete data sets of all sites for enhanced source apportionment.A total of nine different factors were resolved (notable elements in brackets): in PM10-2.5, brake wear (Cu, Zr, Sb, Ba), other traffic-related (Fe), resuspended dust (Si, Ca), sea/road salt (Cl), aged sea salt (Na, Mg) and industrial (Cr, Ni); in PM2.5-1.0, brake wear, other traffic-related, resuspended dust, sea/road salt, aged sea salt and S-rich (S); and in PM1.0-0.3, traffic-related (Fe, Cu, Zr, Sb, Ba), resuspended dust, sea/road salt, aged sea salt, reacted Cl (Cl), S-rich and solid fuel (K, Pb). Human activities enhance the kerb-to-rural concentration gradients of coarse aged sea salt, typically considered to have a natural source, by 1.7-2.2. These site-dependent concentration differences reflect the effect of local resuspension processes in London. The anthropogenically influenced factors traffic (brake wear and other traffic-related processes), dust and sea/road salt provide further kerb-to-rural concentration enhancements by direct source emissions by a factor of 3.5-12.7. The traffic and dust factors are mainly emitted in PM10-2.5 and show strong diurnal variations with concentrations up to 4 times higher during rush hour than during night-time. Regionally influenced S-rich and solid fuel factors, occurring primarily in PM1.0-0.3, have negligible resuspension influences, and concentrations are similar throughout the day and across the regions.
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