Geophysical Research Abstracts Vol. 19, EGU2017-11902, 2017 EGU General Assembly 2017 © Author(s) 2017. CC Attribution 3.0 License.



Spatial variability and sources of ammonia in three European cities

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For the assessment of ammonia (NH3) effects on ecosystems and climate, one would ideally know the emission sources and also the spatial distributions. Agriculture is the largest global source of NH3. However traffic, especially gasoline vehicles, biomass burning or waste management can be significant in urban areas.

Ambient NH3 measurements using cavity ring-down spectroscopy were performed online at high time resolution on a moving vehicle in three cities: Zurich (Switzerland), Tartu (Estonia) and Tallinn (Estonia). Initial tests showed that a regular inlet cannot be used. A heated line including an auxiliary flow was finally deployed to minimize NH3 adsorption onto the inlet walls. We will present the characterization of the response and recovery times of the measurement system which was used to deconvolve the true NH3 signal from the remaining adsorption-induced hysteresis. Parallel measurements with an Aerodyne aerosol mass spectrometer were used to correct the observed NH3 for the contribution of ammonium nitrate (NH4NO₃) which completely evaporated to NH3 and nitric acid (HNO₃) in the heated line at the chosen temperature, in contrast to ammonium sulfate. Finally, quantitative measurements of ambient NH3 are possible with sufficient time resolution to enable measurement of NH3 point or line sources with a mobile sampling platform.

The NH3 analyzer and the aerosol mass spectrometer were complemented by an aethalometer to measure black carbon and various gas-phase analyzers to enable a complete characterization of the sources of air pollution, including the spatial distributions and the regional background concentrations and urban increments of all measured components. Although at all three locations similar urban increment levels of organic aerosols were attributed to biomass burning and traffic, traffic emissions clearly dominated the city enhancements of NH3, equivalent black carbon (eBC) and carbon dioxide (CO₂). Concentration gradients in areas strongly influenced by traffic emissions (including drives in and out various tunnels) were used to determine fleet average emission factors (EF) for the traffic-related pollutants. Significant differences were found between the EFs of certain components in the three cities, which were to some degree consistent with an older vehicle fleet in Estonia compared to Switzerland. Using the determined EFs we show that traffic can basically fully explain the NH3 increments in the three cities and also a non-negligible fraction of the background concentrations, which are known to be mostly related to agricultural activities. Comparisons to a European emission inventory will be discussed.