Summer ammonia measurements in a densely populated Mediterranean city

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Atmospheric ammonia (NH_3) is among the most abundant nitrogen compounds in the atmosphere and it plays an important role in: a) neutralisation of atmospheric acids to form ammonium salts, as ammonium sulfate ($(NH_4)_2SO_4$) and ammonium bisulfate ($(NH_4)HSO_4$) from sulfuric acid, ammonium nitrate (NH_4NO_3) from nitric acid and ammonium chloride (NH_4Cl) from hydrochloric acid, and b) eutrophication and acidification of the ecosystem by nitrogen (N) deposition (Flechard *et al.*, 2011). Overall, agriculture is estimated to contribute for 94% to the total NH_3 emission in Europe with livestock being the largest category in its emission inventory (EEA Report, 2011). Other sources of NH_3 include animal excreta, biomass burning, industries, coal burning, human breath, sweat and smoking, pets, sewage systems, wastes and vehicle emissions (Sutton *et al.*, 2000). Globally these sources form a minor part of the emissions but they might be relevant locally playing the greater role in ammonia emissions. At urban level for example there is a growing concern related with the emissions of ammonia after the introduction of gasoline-powered vehicles equipped with three-way catalytic converters and diesel-powered vehicles adopting selective catalytic reduction (SCR) system.

Measurements of ambient concentrations of gas-phase ammonia were performed in Barcelona (NE Spain) in summer between May and September 2011. Two measurement sites were selected: one in an urban background traffic-influenced area (UB) and the other in the historical city centre (CC). Levels of ammonia were higher at CC (5.6 ± 2.1 μg/m^3 or 7.5 ± 2.8 ppbv) compared with UB (2.2 ± 1.0 μg/m^3 or 2.9 ± 1.3 ppbv). This difference is attributed to the contribution from non-traffic sources such as waste containers and open markets more dense in the densely populated historical city centre. Under high temperatures in summer these sources had the potential to increase the ambient levels of ammonia well above the urban-background-traffic-influenced UB measurement station. The levels of NH_3 measured in Barcelona, especially high in the old city, may contribute to the high mean annual concentrations of secondary sulfate and nitrate measured in Barcelona compared with other cities in Spain affected by high traffic intensity. In Madrid (around 3 millions inhabitants) for example considerably lower concentrations of NH_3 have been observed compared with Barcelona. These high NH_3 concentrations in Barcelona may explain the increase of around 4 μg/m^3 in the mean annual concentrations of fine PM ($PM_{2.5}$) measured in Barcelona compared with Madrid. Finally, the concentrations of ammonia measured in Barcelona may also be the reason for the nucleation episodes involving NH_4HSO_4 formation which have been observed in Barcelona and which caused high levels of ultrafine particles (Reche *et al.*, 2011).

Ancillary measurements, including PM_{10}, $PM_{2.5}$, PM_1 levels (Particulate Matter with aerodynamic diameter smaller than 10 μm, 2.5 μm, and 1 μm), gases and black carbon concentrations and meteorological data, were performed during the measurement campaign. The analysis of specific periods (3 special cases) during the campaign revealed that road traffic was a significant source of NH_3. However, its effect was more evident at UB compared with CC where it was masked given the high levels of NH_3 measured in the old city. The relationship between SO_4^{2-} daily concentrations and gas-fraction ammonia ($NH_3/(NH_3+NH_4^+)$) revealed that the gas-to-phase partitioning (volatilization or ammonium salts formation) also played an important role in the evolution of NH_3 concentration in summer in Barcelona.

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