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## Changes of ammonia and nitrogen oxide emissions in South-eastern Europe inferred from an Elbrus (Caucasus, Russia) ice core record (1774-2009 CE)

### Content

With the aim to reconstruct the history of atmospheric nitrogen pollution in south-eastern Europe, the ice core records of ammonium and nitrate covering the years 1774-2009 were extracted from a 182 m long ice core drilled in 2009 at Mount Elbrus in the Caucasus, Russia.

The Elbrus ammonium ice-core record indicates a 4-fold increase from pre 1780 to 1990-2010. In summer, when both the vertical atmospheric mixing and anthropogenic ammonia emissions dominated by agriculture sector are strengthened, a moderate increase of ammonium already took place from 1774 and 1880. The summer ammonium increase accelerated a first time at the beginning of the 20th century, and a second time after 1950, levels reaching a plateau between 1980 and 2010. This ice-core trend is compared to estimated past anthropogenic NH<sub>3</sub> emissions in Europe by using state-of-the-art atmospheric transport (FLEXPART model driven with 0.5°x0.5° ERA-5 reanalysis data) simulations of submicron aerosol. It is shown that the ammonium Elbrus ice core trend is in excellent agreement with estimates of past NH<sub>3</sub> emissions from south-eastern Europe (South European Russia, Turkey, southern Ukraine). The ammonium level of ice deposited prior to agricultural activities is found to represent ~11-15% of the 1980-2010 level. This value is in the lower range of estimates at the global scale of the contribution of natural ammonia emission to total (natural plus anthropic) ammonia emissions.

A potentially existing trend of nitrate in the Elbrus ice core remained insignificant prior to ~1945. The well-marked 3-fold increase seen in the Elbrus ice core between 1950 and 1980 followed by a stabilization of levels between 1980 and 2010 is consistent with estimated past emissions of NO<sub>x</sub> from south-eastern Europe. In agreement with the partitioning at the global scale of natural and anthropogenic emissions of NO<sub>x</sub>, nitrate levels in Elbrus ice deposited prior to 1940 represent 26% of those deposited at the end of the 20th century.

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