

200 years of NH₃ inventory in Europe inferred from ¹⁵N of NH₄⁺ in Mont-Blanc ice core with unexpected recent combustion-related emissions

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Today, anthropogenic emissions of ammonia (NH₃) exceed natural emissions causing substantial impacts on radiative forcing, ecosystems, and biodiversity (Sutton et al., 2011). To mitigate these consequences and their associated costs, NH₃ abatement is becoming a priority (Gu et al., 2021). Existing European NH₃ inventories suggest emissions from fertilizers and animal manure to be the main NH₃ source through the last decades (European Environment Agency, 2021). However, a growing body of evidence argues against presumed proportions of sources in NH₃ inventories (Chen et al., 2022).

Ice cores are well-suited archives of past atmospheric composition. We developed a method for extracting NH₄⁺, the deposited form of NH₃, and analysing its ¹⁵N isotopic composition in ice cores. Applying it to an ice core from the Col Du Dôme glacier (CDD, 4250 m asl, Mt-Blanc), we can reconstruct NH₃ changes regarding its atmospheric reactivity and its emission sources since the preindustrial era in Europe.

Here we present the history of δ¹⁵N(NH₄⁺) in the CDD ice core the offering first inventory NH₃ sources in Western Europe for 200 years. During the 19th century, NH₃ emissions decreased from an agricultural share of 60% to 35% by 1900. From 1900 onwards, the evolution of agricultural techniques certainly explains the measured increase in agriculture-induced emissions until the 1950s.

After 1950s, a large increase of NH₄⁺ concentration in the ice is accompanied with higher combustion-related NH₃ contribution. For 2010-2016, only 38% of total emissions is explained by agriculture, in strong disagreement with the EMEP inventory (95%).

Our findings demonstrate that the agriculture emission abatement policies have had substantial results in diminishing its contribution to NH₃ emissions while, the Euro5 and Euro6 policies unintendedly conducted to NH₃ production to such extent that it can become the main source of NH₃ in city centres (Elser et al., 2018).

In a desire to control air quality, our measurements show that public policies must now support a reduction in NH₃ emissions from vehicular, industrial and biomass combustion.

