

Reviewer 2.

The manuscript presents a thorough analysis of ammonia (NH₃) emissions over several centuries using an ice core record from Mount Elbrus. It focuses on both natural and anthropogenic sources of ammonia in the atmosphere and emphasizes the critical sources from agriculture. The study is significant as it provides insights into historical emission trends and their environmental impacts, contributing valuable data to atmospheric and environmental science, especially in the context of increasing agricultural practices and climate change. The proposed methodology and results in this paper are commendable and will undoubtedly serve as a reference point for future research in the field. Here are my specific suggestions.

We would like to sincerely thank the reviewer for the thoughtful comments, which have helped us improve and clarify the manuscript.

The introduction is generally clear, it might benefit from a brief overview of the significance of ammonia emissions in the context of atmospheric science and environmental policy to set a stronger foundation for the research.

Thank you for the suggestion. We agree that highlighting the significance of ammonia emissions is important. In fact, we have already addressed this in the introduction, where we discussed the role of ammonia in atmospheric chemistry and its environmental impact: "Gaseous ammonia is the most abundant alkaline gas in the atmosphere and represents a major component of total reactive nitrogen. It plays an important role in determining the overall acidity (alkalinity) of precipitation. A large portion of atmospheric aerosols, acting as cloud condensation nuclei, consists of sulfate neutralized to various extents by NH₃. Ammonia and ammonium (collectively abbreviated as NH_x) are key nutrients that fertilize plants. Too large inputs of N to the environment may, however, lead to eutrophication of terrestrial and aquatic ecosystems and thus threaten the biodiversity (Asman et al., 1998; Galloway et al., 2003). Therefore, the growing NH₃ emissions resulting from fertilization applied to meet the need to sustain food production for a growing human population impact the environment."

We believe this provides a strong foundation for the significance of ammonia emissions in the context of atmospheric science and environmental policy.

The methodology for the comparison process in this paper, including statistical methods or software tools employed, and any adjustments made for the comparison should be clearly outlined. Ensuring that the ice core data, model input files, and emission inventories are publicly available would greatly enhance reproducibility.

Providing access to these datasets through repositories or as supplementary materials is important. This can ensure that other researchers can replicate the findings reliably.

Thank you for the suggestion. The methodologies used in this paper as recognized in your next comment are clearly outlined in the text. We did not apply specific statistical methods for the comparison process.

Regarding software tools, section 2.2 already details that past NH₄⁺ deposition fluxes at the ELB site were calculated by weighting past NH₃ emissions from each grid cell of the inventory by its emission sensitivity and summing over all grid cells to obtain the simulated deposition rate. These calculations were performed using the CDO (Climate Data Operator) software.

As for emission inventories, we used the global dataset of anthropogenic NH₃ emissions (Hoesly et al., 2018), presented in NetCDF format with a 0.5° × 0.5° spatial resolution. This dataset is

publicly available (see Figure 2 caption, Hoesly et al., 2018: <https://github.com/JGCRI/CEDS/>), so there is no need to replicate it here.

We are unable to provide access to the meteorological input data used in the model simulations, as these datasets amount to hundreds of terabytes and we do not have permission from ECMWF to share them.

Lastly, as mentioned in the "Data Availability" section, ammonium concentration data can be accessed at <https://zenodo.org/records/12549687> (Legrand et al., 2024).

Although the methods are described in detail, some sections could use additional clarity, particularly for readers less familiar with specific techniques. It is recommended to supplement the text with more detailed explanations of these methodologies and any assumptions made during the analysis. Consider adding diagrams or flowcharts to visually represent the process.

Thank you for your valuable feedback. Although the methods are described in detail. Data discussed in this paper (mainly ammonium and sulfate) were obtained using the well-known ion chromatography. There is no specific assumption made behind such routine measurements. The reader can find detailed working conditions in earlier publications as referenced here (Preunkert and Legrand, 2013, Legrand et al., 2013). In the revised version, however, we have also included the following statement *"the detection limit for ammonium is close to 1 ng g⁻¹ so remaining well below mean (low) winter concentrations that typically ranged from 10 to 20 ng g⁻¹."*

It is suggested to expand certain sections of the discussion to provide a more in-depth analysis of the implications of the findings, particularly expanding the discussion to cover the broader implications of the findings, potential limitations, and areas for future research would be beneficial.

Thank you for your insightful suggestions. We recognize the importance of expanding certain sections of the discussion to provide a more comprehensive analysis of the implications of our findings. In the conclusion, we have already emphasized that "Transport-chemistry model simulations are welcome to further evaluate NH₄⁺ ice core records; however, they would require consideration of increasing SO₂ and NO emissions, as well as dust aerosol and its heterogeneous interactions with acidic species and NH₃."

Additionally, we have now included the following statement: "The ice-core trends are less documented for winter than for summer. A better understanding of past ammonium changes in winter motivates the search for another glacier site in the Caucasus that may provide better preservation of winter snow (due to less wind erosion)." We believe these additions help to highlight potential areas for future research.