

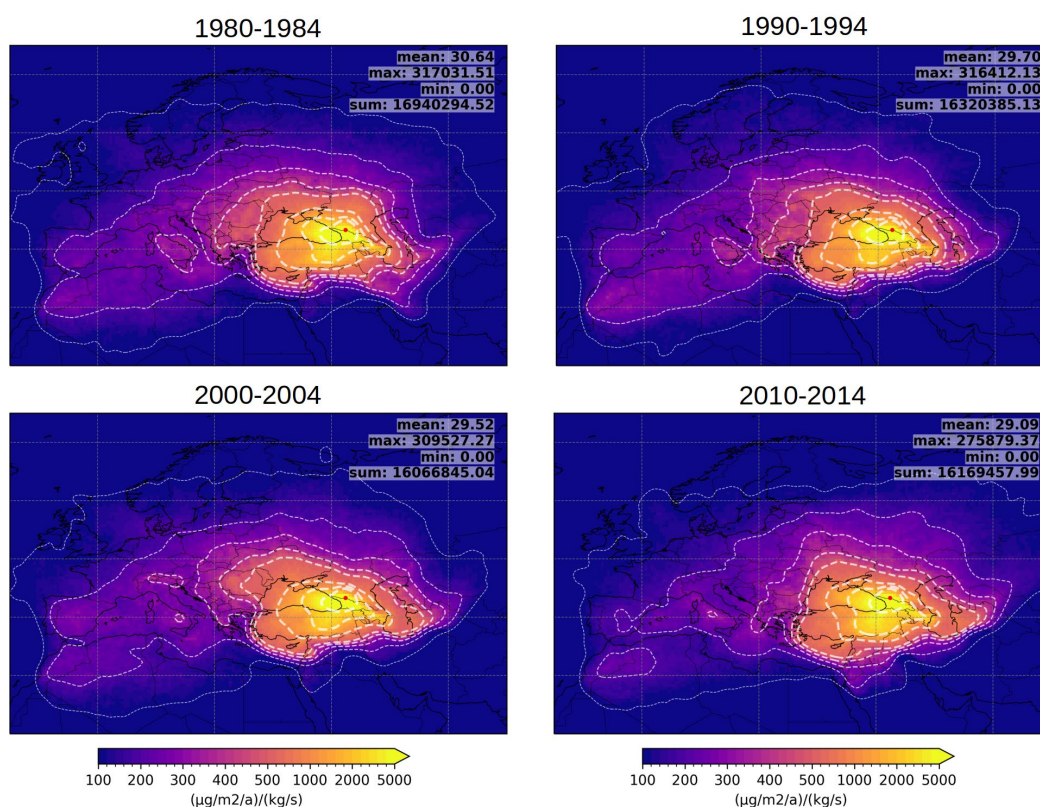
### Reviewer 3.

The authors investigate historical atmospheric ammonia (NH<sub>3</sub>) pollution using a 182 m long ice core from Mount Elbrus. The ice core data reveal a 3.5-fold increase in ammonium (NH<sub>4</sub><sup>+</sup>) concentrations from approximately 1750 to 2009, with a significant rise post-1950 due to industrial and agricultural activities. The study utilizes FLEXPART atmospheric transport modeling to compare the ice core trends with past anthropogenic NH<sub>3</sub> emissions, highlighting the substantial impact of human activities on atmospheric NH<sub>3</sub> levels. The research also differentiates between natural and anthropogenic contributions to NH<sub>4</sub><sup>+</sup> concentrations, providing a baseline for pre-industrial natural emissions and underscoring the predominance of agricultural emissions in recent decades. The authors are leaders in this type of work; the data and methodology are all sound; and the topic and scope will be of interest to ACP readers. Overall, this article makes a significant contribution to the understanding of historical ammonia emissions in south-eastern Europe. Its robust dataset, interdisciplinary approach, and detailed methodology are commendable. I believe the paper is publishable mostly as is, but I encourage the authors to consider these points when revising:

[Thank you for your thoughtful review.](#)

**Model Assumptions and Limitations:** The study relies heavily on the FLEXPART model, which, while robust, has limitations. The assumption that atmospheric transport has not changed significantly over the ice core record period might oversimplify complex atmospheric dynamics. Some type of sensitivity analyses to explore the impact of varying transport conditions would be helpful. [Thank you for your valuable feedback regarding the FLEXPART model and its limitations. We agree that the assumption of constant atmospheric transport over the ice core record may oversimplify the complexities of atmospheric dynamics. In response, we have now included an analysis of the temporal variability of emission sensitivities over the 1980-2019 period. As shown in the figure added to the Supplementary Information \(see below\), the emission sensitivities did not change significantly during this period.](#)

## Variability of summer emission sensitivity – Mt. Elbrus



**Winter Data Uncertainties:** The ice core data for winter months are less reliable due to fewer samples and potential wind erosion. This limitation weakens the study's conclusions about seasonal variations in NH<sub>3</sub> emissions. Additional measures or methods to improve winter data accuracy would strengthen the overall findings. *Thank you for highlighting the limitations of the ice core data for winter months, including fewer samples and potential wind erosion. We agree that these factors can weaken the study's conclusions regarding seasonal variations in NH<sub>3</sub> emissions.*

*While it is currently not feasible to implement additional measures to improve winter data accuracy, we have added the following statement in the conclusion: "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 this addition emphasizes the need for further investigation in this area.*

**Spatial Resolution of Emission Sources:** The study identifies significant contributors to NH<sub>4</sub><sup>+</sup> deposition, but the spatial resolution of these sources could be improved. A finer resolution might reveal more localized sources and patterns of emissions, offering better-targeted mitigation strategies. *We agree that a finer resolution could provide a more detailed understanding of localized sources and emission patterns, ultimately leading to better-targeted mitigation strategies. In this study, we utilized emissions data available at a resolution of 0.5° × 0.5°, which aligns with the resolution of the FLEXPART simulations. This resolution allows us to identify "hot spots" in several regions, such as Brittany in France, northern Italy, and western Ukraine (see Figure 2).*

*We appreciate your suggestion, as it highlights an important consideration for future research.*

**Consideration of Other Pollutants:** While the focus on NH<sub>3</sub> is clear, the interplay between NH<sub>3</sub> and other atmospheric pollutants (e.g., SO<sub>2</sub>, NO<sub>2</sub>) is mentioned but not deeply explored. A more detailed examination of how these pollutants interact and affect NH<sub>3</sub> deposition could provide a fuller picture of atmospheric chemistry dynamics.

*We agree that a more detailed examination of these interactions could provide a fuller understanding of atmospheric chemistry dynamics. However, NH<sub>3</sub>-NO<sub>x</sub>-SO<sub>2</sub> interactions are quite complex and not always fully understood. As concluded in the paper, further progress in this area would require the deployment of a comprehensive transport-chemistry model that includes a complete description of SO<sub>2</sub>, NO<sub>x</sub>, and NH<sub>3</sub> chemistry, as well as the effects of historical dust changes. Unfortunately, such a model is not currently available.*

**Climate Change Implications:** The study briefly touches on the potential impact of climate change on NH<sub>3</sub> emissions but does not delve deeply into future projections. Integrating climate models to predict future NH<sub>3</sub> emissions under different climate scenarios would add valuable forward-looking insights.

*Thank you for your insightful comment. We agree that integrating climate models to predict future NH<sub>3</sub> emissions under various climate scenarios would provide valuable insights. However, as with your previous comment, discussing future projections is challenging without the use of transport-chemistry model simulations that account for anticipated changes in SO<sub>2</sub> and NO<sub>2</sub> emissions, as well as dust aerosol.*

*In response to both your comments we have noted in the conclusion that “Transport-chemistry model simulations are welcome to further evaluate NH<sub>4</sub><sup>+</sup> ice core records; however, they would require consideration of increasing SO<sub>2</sub> and NO emissions, as well as dust aerosol and its heterogeneous interactions with acidic species and NH<sub>3</sub>.”*