Reviewer 1.

This study investigated that the historical levels of atmospheric ammonia (NH_3) pollution in south-eastern Europe. The results showed that The NH^{4+} ice core record indicates a 3.5-fold increase of annual concentrations from 34 ± 7 ng g^{-1}(~1750-1830) to 117 ± 23 ng g^{-1} over the recent decades (1980-2009). And this pre-1750 natural level mainly related to natural soil emissions represents ~20% of the 1980-2009 NH^{4+} level, a level mainly related to current agricultural emissions that almost completely outweigh biogenic emissions from natural soils. I recommend the manuscript be revised before being accepted for publication.

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

Line 50, the authors mention: "up to now only two studies have compared ammonium trends extracted from Alpine ice cores to atmospheric chemistry-transport models (Engardt et al., 2017; Fagerli et al., 2007)", but what are the key findings in these previous studies? A brief introduction to previous studies can better highlight the research content.

These two studies were limited to past changes having occurred over the 20th century whereas anthropogenic NH_3 emissions were already significant during the 19th century. Also, the two studies were conducted at glaciers essentially impacted by western European emissions. In the revised version we added:

"Such studies remain scarce, and up to now only two studies have compared ammonium trends extracted from Alpine ice cores to atmospheric chemistry-transport models (Engardt et al., 2017; Fagerli et al., 2007) with the aim to constrain past ammonia emissions in western Europe over the 20th century. The short lifetime of atmospheric aerosols (days to weeks) and the regional character of ammonia emissions, however, motivated further studies conducted in other regions and extending back to the 19th century when agricultural activities started to be significant."

Line 70, "a difference by ~25 years is observed at the end of the record". Does this difference have an impact on the comparison of results from analyzing observations and simulations?

We recognize that, as it reads, the text was misleading. First, we now clarify that in this study we used the most recent dating that was established with more information than the previous one. Whereas there is indeed a difference of 25 years between the two dating around 1750, that does not represent a dating uncertainty. Instead, as now better explained in the text, the uncertainty on the depth of the Tambora layer (153.7 m or 154.7 m depth) leads to an uncertainty of 4 years around 1815 for the new dating. As discussed in section 5.1, the ammonium concentration increase remained limited to 0.1% yr^{-1} between 1750 and 1850 compared to 2.8% yr^{-1} after 1950, implying that dating uncertainty in the bottom part of the record would not significantly impact the discussion of the temporal trend of the last 250 years. The text has been reworded as :

"Based on complementary data including the acidity, the dating was recently revisited by Mikhalenko et al. (2024), suggesting the presence of the 1815 Tambora horizon either at 153.7 or 154.7 m depth and an age of CE 1752 ±4 years at 168.6 m depth. This more accurate dating was used in this study, the uncertainty around 1815 CE being of 4 years. Note that as discussed in section 5.1, the ammonium concentration increase remained limited between 1750 and 1850 compared to the post 1950 period, implying that dating uncertainty in the bottom part

of the record does not significantly modify discussions on the main changes that had occurred over the two last centuries."

Lines 73-74, "removing ~3 mm with a pre-cleaned electric plane tool under a clean air bench.", What is the scientific basis for it, please add.

This is a rather standard decontamination technique described and justified before including the paper cited already in the text (Preunkert and Legrand; 2013). We do not think that further explanation is necessary for this.

That ensures that the outer part of piece of ice (often contaminated) was removed permitting to obtain free-contamination piece of ice, as previously successfully tested for Greenland (Fischer et al., 1998), Alpine (Preunkert et al., 2001; Preunkert and Legrand; 2013), and Caucasus ice (Preunkert et al., 2019).

Fischer, H. , Wagenbach, D. and Kipfstuhl, S. (1998): Sulfate and nitrate firn concentrations on the Greenland Ice Sheet 1. Large-scale geographical deposition changes, Journal of Geophysical Research D17, 103 , pp. 21927-21934

Preunkert, S., M. Legrand, D. Wagenbach, and H. Fischer, Sulfate trends in a Col du Dôme (French Alps) ice core : A record of anthropogenic sulfate levels in the European mid-troposphere over the 20th century, *J. Geophys. Res.*, 106, 31,991-32,004, 2001.

Line 259, "countries for which FLEXPART simulations indicate a significant contribution to the NH^{4+} deposition in ELB ice include Russia and several Middle East countries (Turkey, Iran, Egypt, Fig.4b)". How this "significant contribution" is judged, the graph shows that the rest of the countries are relatively high compared to Iran or Egypt.

Taken into account, first we reworded this sentence

"On the other hand, countries for which FLEXPART simulations indicate a significant contribution to the NH_4^+ deposition in ELB ice include the former USSR (Russia, Ukraine, and Georgia) and the Middle East (Turkey) (Fig. 4b) for which agricultural areas are documented since 1600 CE (https://ourworldindata.org/grapher/total-agricultural-area-over-the-long-term), data being, however, not detailed enough for weighting emissions from the different countries to the NH_4^+ natural level at the ELB site."

Second, concerning other countries we added in the next paragraph: "*These deposition flux changes reflect past emission changes characterized by a growth of NH_3 emissions that took place after World War II in many countries with major contributions from Russia, Turkey, Georgia, and Ukraine. As seen in Fig 4b, even with weakened emission sensitivities (Fig. 1), due to large NH_3 emissions (Fig. 2), other countries located further west such as Bulgaria, Albania, Hungary, Macedonia, part of Italy, Hungary, Slovakia, and Czech Republic still significantly contribute to the deposition at the ELB site. Furthermore, the maximum of emissions that took place in the late 1980s, during the perestroika in USSR, is also well recorded in ELB summer ice. As a result of decreasing NH_3 emissions in Russia and Ukraine in 1988-1989 (Fig. 2), Turkey became in the 1990s the main contributor of NH_4^+ deposition at the ELB site (Fig. 4b)."*

Fig.6b and Fig.6d have a low correlation for the scatter fit (0.26 and 0.37), can the authors try a segmented fit, which is negatively correlated up to the first half of the x-axis as can be seen in the figure.

Taken into account. We do not think that this will help here, the scattering being too high, likely due to low winter values together with a poor representativeness of the thin winter ice layers that characterized the bottom part of the ice core which is stated in the text.

Lines 374-376, "increased modestly between 1900 and 1950". That's too vague a descriptionare. It would have been clearer if a comparison in terms of data had been given. This makes the paper more rigorous and academic. Note other similar descriptions in the manuscript.

Taken into account, we now specify "In summer, concentration and deposition of SO_4^{2-}red remained quasi-unchanged from 1750 to ~1900 CE, increased modestly at a rate of 3.5% yr^{-1} between 1900 and 1950. The increase then accelerated until ~the middle of the 1980s (12% yr^{-1}) followed by a strong decrease from the early 1990s to 2009 (11% yr^{-1}, Fig. 11)."

Lines 400-415, "In winter, $SO4^{2-}$red observed deposition fluxes in ice deposited … would permit to better evaluate this effect". The authors explain a lot about this phenomenon, but there doesn't seem to be a clear explanation, the authors should summarize and analyze to get a clear point of explanation.

Taken into account.

It seems to us that it was stated in the draft as "*The difference between observed and simulated depositions are similar for sulfate and ammonium and we can therefore rule out that NH_3 emissions in winter were overestimated. Rather, these differences may be a result of deficiencies in the FLEXPART simulations, e.g., related to an under-representation of winter-time atmospheric inversion layers in the meteorological input data which would hinder vertical transport, or an underestimation of the observed winter deposition due to winter snow being blown away at the ELB site.*"

We now add a sentence in the conclusion on this point: "*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 possibly experiences a better preservation of winter snow (less wind erosion).*"

The manuscript is too long and not clear enough. The authors should adjust it so that the structure of the manuscript is expressed more clearly and concisely.

We appreciate the reviewer's comment but would like to clarify that we are a bit uncertain about the suggestion to shorten or reorganize the manuscript. The other two reviewers did not recommend any structural changes. In fact one suggested expanding the discussion section while the other recommended publishing the manuscript as is. Additionally, the ACP journal does not provide explicit guidelines on manuscript length. However, in response to the feedback, we have reduced the number of figures in the main text, moving two to the Supplementary Information as recommended.

Line 23, "0.5°x0.5°" should to be multiplication sign "×", not the letter "x". Note the change!

Thank you for pointing this out. We acknowledge the convention of using the multiplication sign "×" instead of the letter "x". However, in our previous publications in JGR and GRL, as well as commonly in ACP papers, the "x" format (e.g., 5° x 5°) has been used. We will review this point carefully during the proof editing stage.

Now that there are too many graphs in the manuscript, please organize some of them and place them in the supplementary Material.

Thank you for your suggestion. While the journal does not impose limitations on the number of figures, we agree that streamlining the manuscript would be beneficial. In response, we have moved two figures to the Supplementary Information (SI): Figure 7 (Past changes of the agricultural area) and Figure 4b. Additionally, we have combined Figure 4a with Figure 9 to reduce redundancy and improve organization.