## Response to reviewers and description on the revised manuscript

First, we thank the reviewers and editor for taking the time to review our work. We appreciate the constructive comments made to improve the manuscript.
The manuscript has been thoroughly and carefully revised in line with the evaluations received.

Our point-by-point responses (in magenta, unformatted text) following the referee's comments (in black) can be found below.

The previous text version is in blue and corrections applied to the manuscript appear in **bold magenta**.

Please note that the lines mentioned refer to the newly submitted version.

**Reviewer 2:**

In "Evaluating present-day and future impacts of agricultural ammonia emissions on atmospheric chemistry and climate", the authors present simulations of present day and future ammonia pollution at a global scale with the LMDZ-INCA model with new and improved NH3 emissions from ORCHIDEE, then evaluate the impact of future ammonia burden on air quality and climate. Comparison of present-day simulated NH3 with satellite and ground-based measurements shows improvement from other inventories. Future projections are informed by SSP scenarios, and consider both changes in aerosol burden and composition, as well as changes in N2O production from NH3 oxidation. Overall, this manuscript presents an interesting outlook on future air quality and climate conditions from the perspective of ammonia emissions that is within the scope of ACP. I recommend publication following some clarification and revision of methods and results.

We are thankful for the constructive comments given by Reviewer 2. Our point-by-point responses can be found below.

General Notes:

When comparing against ground-based data, no consideration appears to have been made for differences in monitor spatial or temporal coverage, which may hinder direct comparisons. For example, it appears ground-based measurements are only reported for 2015, which is mismatched temporally with the model simulation.

We fully agree with the reviewer that the mismatch in the years of the model and the observations was not optimal for a consistent evaluation.

To address this important point we extended the CAMEO simulation until 2015 and compared the results against the same year of observations. We replaced the Figures 4 to 6 in the revised manuscript and adjusted Table 4 and text accordingly.

In addition, as also requested by Reviewer 1, the comparison to 2010 has been added to the Supplementary Material. The results for 2010 appear less robust than in 2015 due to much fewer observation numbers as highlighted in the following table with the example for $NH_3$ observations:

| # obs | EMEP cc | UK Networks | NNDM | EANET | US EPA | NAPS |
|---|---|---|---|---|---|---|
| 2010 | 26 | 23 | 10 | 25 | 11 | 7 |
| 2015 | **38** | 22 | **25** | 27 | **31** | 7 |

This new sentence has been incorporated:
**An evaluation for 2010 has also been conducted to enhance the robustness of our findings and similar regional signals are found as for 2015.**
**Owing to the fewer observations available globally compared to 2015, these results are presented in the Supplementary Material (Fig. S7, S8, and S9).**

What averaging methods are employed to gather a single value for different species that are each measured with different methods and temporal resolution? Beyond data completion of data record, is any further QA performed with this data to ensure measurement validity?
The spatial matching between model-observation has been performed with a simple data 'mining' corresponding to the extraction of the closest pixel for each network site. The spatial averages of each network are then performed to gather a single value for different species. Regarding the temporal coverage, we ensured that the yearly period was covered at 75% in the observation dataset for each site and performed yearly averages in the model data.
For the present study, no further QA has been performed on the observation data but we acknowledge that the quality of the measurement is a crucial aspect that can bring uncertainties to the evaluation. By using both satellite and ground-based measurements, we hope to have demonstrated that the model produces relatively reasonable results.
Precision has been incorporated into the manuscript:
**As recommended in Ge et al., 2021, we only consider measurements where 75% of the year was captured to avoid bias in our analysis and we perform yearly averages on the model data.**

The authors report excellent agreement between measured NH3 and modeled results in the Mid-US in Figure 6; however, there are only 2-3 surface sites shown within the "hotspot" region of the Mid-US to make this comparison, and at least one site appears to have the maximum difference threshold between model and measurements. Surface monitor locations are notably sparse in agricultural regions of the US where emissions tend to be high, and similar biases may exist in other regional networks. Please clarify the methods and findings in this section.
We completely agree with the Reviewer on this aspect, however, the original text does not report "excellent agreement" or similar terms:
"In North America, CAMEO reflects a realistic spatial pattern against measurements with high concentrations located in the Mid-US (> 4 µg.m$^3$) and rather low concentrations on the Mid-Atlantic side.
An underestimation of CAMEO is still observable in the Mid-West region (<2 µg.m$^3$)."
To emphasize the scarcity of the data in the intensive agricultural region of Mid-US, we added the following precision:

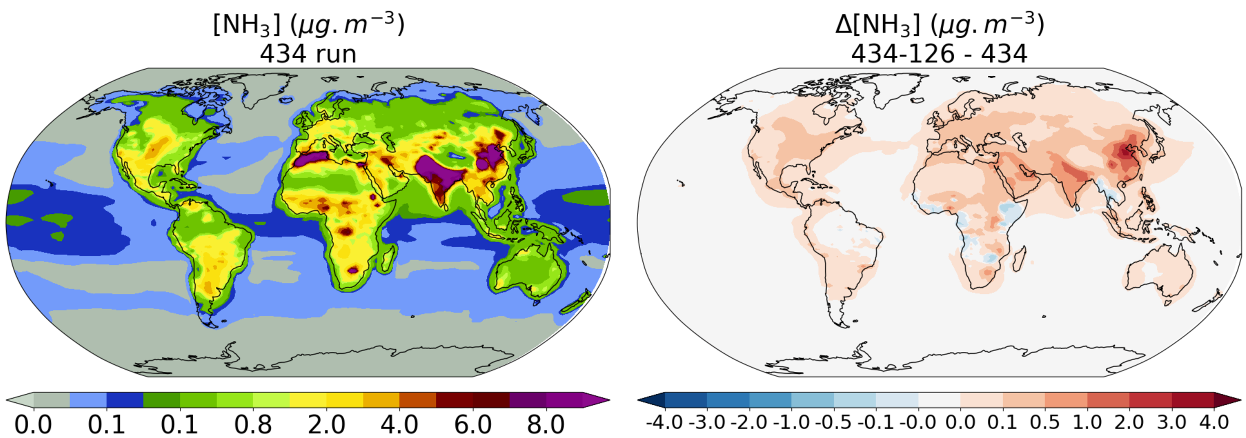
**Even though the spatial gradient is fairly represented in the model, it is crucial to note that only a few observations are available, especially in the Mid-US region, an intensive agricultural area that would benefit from further observation data for more accurate evaluation.**

Further analysis may be warranted regarding the findings of section 5.1, particularly for changes projected in China. This topic is important, but to my knowledge there is no substantial evidence that SO2 and NO2 controls increase ambient ammonia burden, and the authors only reference one publication in this discussion of these results, though others exist for China and other regions. In Warner et al., 2017 GRL, for example, the authors postulate that ammonia increases in China are due to a combination of sulfur controls, increased fertilizer use, and increasing local temperatures. It seems that these variables could be further investigated within the model outputs here.

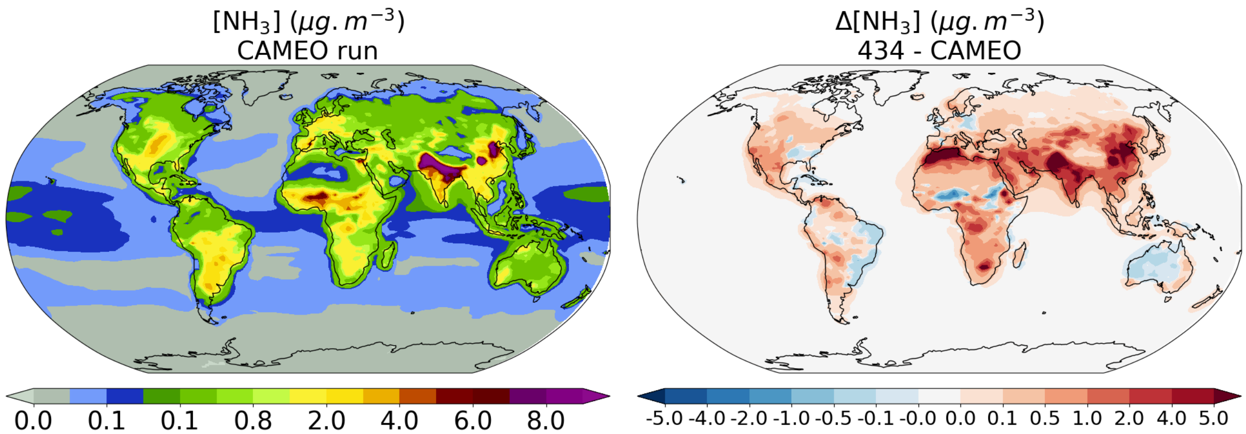
We thank the reviewer for providing an insightful view of the future changes in ammonia burden in intensive agricultural areas such as China.

We think that investigating other variables would be beneficial, however, we want to emphasize that in the framework of our study, the meteorological variables (including local temperatures) are kept at the present-day level. This choice is justified by the fact that we want to isolate the agricultural and industrial drivers that impact atmospheric chemistry.

As highlighted in the simulation experiments by the following Figure, CAMEO434 and CAMEO434-126, we can clearly state that both the decrease of $SO_2$ and NOx are important factors of the increased ammonia burden (2-3 $\mu g.m^{-3}$).




The impact of increased ammonia emissions alone is responsible for at least 5 $\mu g.m^{-3}$ increase in the region (see following figure, already presented in the manuscript), mainly explained by the strong increase in synthetic fertilizer use in the SSP4-3.4 (+30 TgN/yr compared to historical application, see Fig. 4 of https://essopenarchive.org/doi/full/10.22541/essoar.170542263.35872590/v1).




To consider your interesting suggestion we reformulated the paragraph:

**In line with the later study, the effect of the simultaneous reductions in Nox and $SO_2$ emissions in CAMEO[434-126] is even stronger on [$NH_3$] due to the combined increase in $NH_3$ emissions mainly explained by the significant increase in the use of synthetic fertilizers in China (+30 TgN/yr compared to historical application).**

**This is also confirmed by comparing [$NH_3$] from CAMEO[434-126] and CAMEO[434] where $NH_3$ emissions are identical but a slightly stronger impact on the concentrations is highlighted for instance in China and India (Figure 9).**

**It is notable that other combined factors have been shown to significantly contribute to the increased $NH_3$ levels in China.**

**For instance, in Warner et al., 2017, the authors suggest that the rise in ammonia levels in China between 2003 and 2015 can be attributed to sulfur controls, greater fertilizer application, and rising local temperatures.**

**The present study does not explore the impact of meteorological factors, as it focuses on the isolated impact of human-related ammonia emissions.**

I find the expansion of results towards radiative forcing to be a particularly interesting portion of this work; however, the methodologies here are unclear, especially regarding the N2O estimates from ammonia oxidation. These N2O estimates are also highlighted as a major conclusion of this work in the abstract, but the analysis presented in the actual manuscript seems overly brief for this to be a main conclusion. Similarly, the abstract references a range of N2O (0.43 to 2.10 Tg/yr), but only single figures are presented in the results and discussion. This section also refers to figure 5, but N2O is not a component listed within that table. Should changes in the N2O budget as a result of increasing ammonia be a main conclusion of this work, more in-depth analysis should be considered, perhaps with a figure or table dedicated to this section. The conclusion section also presents somewhat different estimates than what is represented in the abstract and main text. It feels startling to see mention and estimates of ammonia/hydrogen economy in this section of the conclusions when it has not been previously mentioned in the manuscript. Clarification of numerical estimates and methodologies in these sections is necessary.

We agree with the reviewer that the results around the $N_2O$ production were lacking context. To do so, the introduction has been completed with a motivation for studying $N_2O$ production:

**The ammonia oxidation pathway mentioned is a direct contributor to nitrous oxide ($N_2O$) emissions in the atmosphere, which is a potent greenhouse gas.**

**Future losses of nitrous oxide could increase significantly due to intensified agricultural emissions and the emerging hydrogen fuel economy, which heavily relies on ammonia as an energy carrier (Hauglustaine et al., 2014, Bertagni et al., 2023).**

The calculation of $N_2O$ production has been added to the Model description section (3) as:

**Ammonia losses occur as a result of both wet and dry deposition, ammonium formation, and the oxidation processes in the gas phase, although the latter contributes only a small amount to its overall loss.**

**However, the loss through this oxidation pathway generates a non-negligible amount of nitrous oxide ($N_2O$).**

**The production of $N_2O$ results from the following reaction:**

**$NH_2 + NO_2 \rightarrow N_2O + H_2O$**

**The overall production rate is calculated as:**

$R_{nh2->N2o} = A \times exp^{-Ea/RT} \times [NH_2] \times [NO_2]$ **with A = 2.1e$^{-12}$ and Ea/R = -650**

This production term is part of Table 5 under the new label "$N_2O$ prod." and is described in the legend as "$N_2O$ production through $NH_3$ gas phase loss (TgN/yr)".

We double-checked the values and corrected them to have consistency between the main text, the conclusions and the abstract.

Specific Comments:

-Line 32: the meteorological variables described are unclear. Is this referring to air temperature and humidity, or soil temperature and moisture?

Indeed, there is a lack of precision. We are primarily referring to soil temperature and moisture. This info has been added in the revised version.

-Line 42: a citation should be used regarding the claim that livestock activities and synthetic fertilizer use are projected to increase.

References have been added.

-Please ensure that citation formatting is consistent throughout the manuscript. For example, on page 2 line 43-48, the authors reference Hauglustaine 2014 on the first sentence but not the second. Later in the same paragraph, the authors reference Beaudor 2024 on both sentences referring to that work.

Thanks, we double-checked that.

-Line 67: clarify the gas-phase and particulate species examined in this work.
We clarified as follows: **trace gases: $NH_3$, $NO_2$ and ionic species: $NH^+_4$ , $NO^-_3$ , $SO_2^{-4}$.**

-Line 68: It may be beneficial to include greater detail of the SSP scenarios used in this work in the introductory section of the manuscript. As mentioned here, it is unclear what is meant exactly by "most and least significant increase".
We agree that more explanation is needed. Therefore additional descriptions have been included in the introduction:
**SSP4-3.4 and SSP5-8.5 describing respectively "A world of deepening inequality", and "Fossil-fueled Development – Taking the Highway" (Calvin et al., 2017, Kriegler et al., 2017), reflect divergent agricultural drivers.**

**In the first place, SSP4-3.4 represents the scenario with the weakest evolution of livestock, while SSP5-8.5 shows the most significant increase among all Shared Socioeconomic Pathways (SSPs) according to Riahi et al., 2017.**
**In line with these trends, the fossil fuel-intensive scenario SSP5-8.5 also experiences the highest demand and production of food and feed crops among the three scenarios considered, as noted by Beaudor et al., 2024.**
**This increase occurs despite low population growth and is driven by the prevalence of diets high in animal products (Fricko et al., 2017, Kriegler et al., 2017).**
**Despite the peak in food and feed crop production, projected fertilizer applications in SSP5-8.5 rise only slightly.**
**This is attributed to the minimal production of biofuel crops, a result of the lack of climate mitigation policies and rapid advancements in agricultural productivity.**
**In contrast, SSP4-3.4 exhibits the highest use of fertilizer and reveals significant regional differences, with high consumption lifestyles among elite socioeconomic categories and low consumption levels for the rest of the population (Calvin et al., 2017).**

Line 72-74: delete "more precisely" and "respectively".
Done.

-Line 75: what is meant by different levels? This type of descriptor is used several times throughout the manuscript but is poorly defined. Delete "structure of the".
The following sentence have been modified in the manuscript for better comprehension:
**In this paper, we present six simulations from LMDZ-INCA, including two present-day simulations, with CEDS and CAMEO inventories for $NH_3$ emissions and four future simulations over 2090-2100 with future $NH_3$ emissions from CAMEO and other sources at different future levels (i.e. globally decreased and regionally-contrasted level of emissions).**

-Page 4: why were the present day years 2004-2014 chosen for this analysis? This time period is somewhat mismatched both with satellite record (2011-2014) and surface-based measurements (2015).

In order to have a more robust signal from the model results, we considered an 11-year period (2004-2014), which has also been selected for future simulations. Even though the CEDS inventory is extended until 2020 (McDuffie et al., 2020), the forcings for the CAMEO model (and ORCHIDEE) are considered a “future period” after 2014. This is why the CAMEO emission coverage influenced our simulation period.
The IASI instrument period selected is justified by the overlap of the two satellites (Metop A and B), which did not offer the best accuracy before 2011.
Regarding the mismatched years with the model and the observations, we extended our historical ammonia emissions with CAMEO to 2015 but the forcings (N input, livestock densities, Land Use maps and meteorology fields) are not considered present-day and it is part of a particular SSP for 2015. We assumed that similar emissions results are expected for any SSP since 2015 is the starting year of all SSPs.

-Lines 97-99: Animal density considerations are an interesting and somewhat underexamined variable towards $NH_3$ emissions, and are described here as a critical input, yet the text does not describe how changing densities are represented. While this is described by a prior publication, that publication is not referenced at this point of the manuscript, and it may be worthwhile to add further detail to the current manuscript.
We agree that more details regarding the livestock density would benefit the manuscript.
We incorporated a description of the methodology of this driver and highlighted the advantages of the use of the CAMEO emission datasets:
**These emission datasets have been recently constructed from a newly gridded livestock product and the use of the global process-based CAMEO before being evaluated against CMIP6 emissions developed by the Integrated Assessment Models (IAMs) in Beaudor et al., 2024.**
**The future livestock distribution has been estimated until 2100, originally, for three divergent SSPs (SSP2-4.5, SSP4-3.4 and SSP5-8.5) through a downscaling method based on regional livestock trends and future grassland areas (the detailed methodology can be found in Beaudor et al., 2024).**
**The simulated agricultural ammonia emissions show a good alignment with the global IAMs estimates of 50 to 66 TgN/yr from CMIP6 under SSP4-3.4 and SSP5-8.5.**
**Although a global agreement is shown between the IAMs and CAMEO future emissions, we identified three interesting advantages in favor of the use of CAMEO emissions:**

- **The consistent consideration of the key ammonia emissions drivers (i.e. N input, meteorology, livestock, and land use) among all future SSPs which is the result of the use of a single process-based model.**
- **The spatial heterogeneity is driven by environmental conditions and not kept constant over time within predefined regions using the information from the historical period.**
- **Incorporating CAMEO into the land component of the IPSL ESM ensures better consistency throughout the various components, including LMDZ-INCA, paving the way for advancements in our understanding.**

Ref: Beaudor, M., Vuichard, N., Lathiere, J., and Hauglustaine, D.: Future trends of agricultural ammonia global emissions in a changing climate,https://doi.org/10.22541/essoar.170542263.35872590/v1, 2024.

-Line 107: change “it represents” to “each represent”
Done.

-Please ensure that all acronyms/abbreviations are described before they are used in the manuscript. For example, LMDZ-INCA is not defined until line 120 but is used several times before this.
Thank you for pointing it out. We checked and defined properly the abbreviation used in the manuscript.

-Line 151: Sander 2015 is an outdated reference and should be updated to Sander 2023.
Thank you for pointing this outdated reference. We corrected it.

-Line 154 & Line 160 : Is there a reason the ERAInterim reanalysis product was used instead of the ERA5 product? What meteorological variables are employed?
In this study, meteorological data from the European Centre for Medium-Range Weather Forecasts (ECMWF) ERA5 reanalysis were used. The relaxation of the GCM winds towards ECMWF meteorology is performed by applying a correction term at each time step to the GCM u and v wind components with a relaxation time of 2.5 h (Hourdin and Issartel, 2000; Hauglustaine et al., 2004). The ECMWF fields are provided every 6 h and interpolated onto the LMDZ grid.
It has been clarified and corrected line 238.

-Line 161: delete “however”.
Done.

-Line 171-172: I’m not fully certain what is meant by low levels and contrasting conditions in these descriptions. It would be helpful to have greater reference towards what each CAMEO-SSP simulation represents, such as an additional column in Table 2 describing textually the purpose of the different conditions.
We thank the reviewer for this suggestion and added a column to Table 2 to explicit the scope of each simulation.

-Line 178: Why are only these years of the IASI record employed for comparison?
The period selected for the IASI comparison has been recommended by the IASI team because of the quality of the retrievals, mainly due to the overlap of Metop B and A satellites.

-Line 203: change to “CAMEO emission also enhance”.
Done.

-Lines 209-214: I find this analysis of compensating errors to be unclear. Where is the 47% bias estimate coming from for the US? From table 3, I see a 46% difference between Mean Obs and MBE CEDS. What is the threshold for significance of these differences?

We agree with the reviewer that this calculation was confusing since the MBE for CAMEO in that particular case is closed to 0.

This sentence has been corrected:

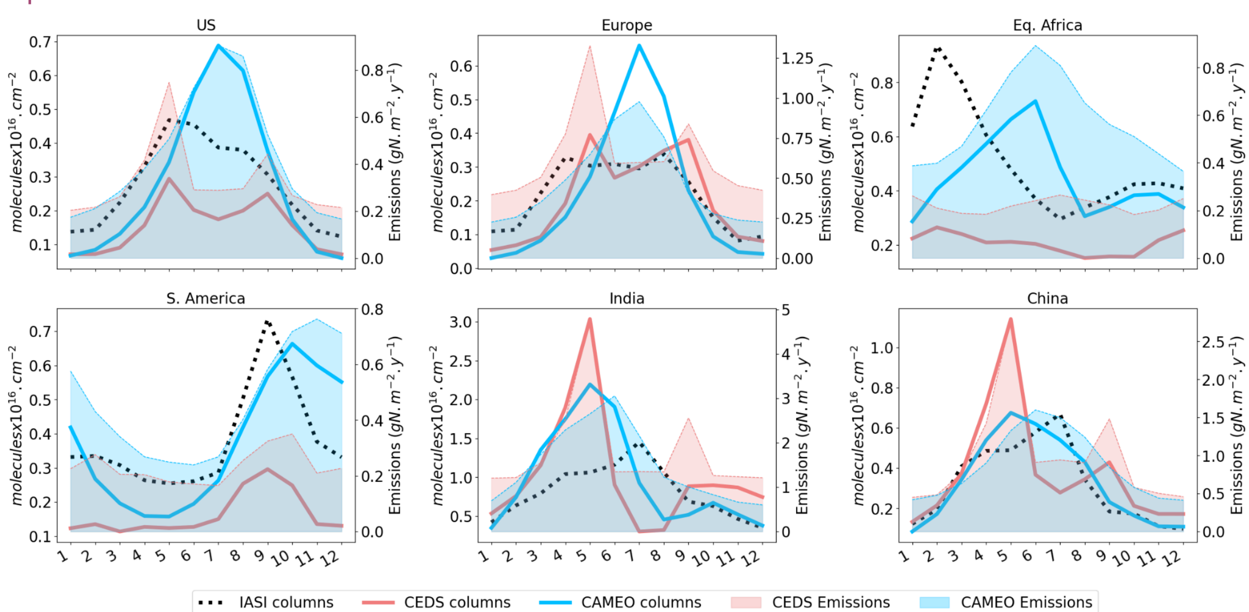
**Using CAMEO also significantly reduced the modeled bias over the US, with an MBE close to 0 (Table3).**

The sentence about the compensating error has been clarified also as follows:

**However, it is important to note potential compensating errors within the regions, particularly in the selected African region (shown in the black box in Figure 1).**
**For instance, in the Saharan area, CAMEO emissions cause an overestimation of column values by 0.3 molecules $10^{16}$ cm$^{-2}$.**
**In contrast, in the tropical Sub-Saharan zone, these emissions lead to an underestimation of column values by -0.4 molecules $10^{16}$ cm$^{-2}$ (-45%).**

-Line 220: Why is the seasonal cycle omitted? Could this be added to the supporting information?

We replaced Figure 3 by the following figure in which the seasonal cycle of the emissions is also represented.




-Page 9: An additional figure showing the difference between CAMEO and CEDS would be helpful, possibly for the Supporting information.

This has been added to Figure S1.

In this figure and others (e.g., Figure 12), differences between outputs are reported only by absolute differences—would it be more clear to represent these as relative (%) differences between outputs?
We considered this suggestion and mentioned some percentage difference values in the text into parenthesis when appropriate. For instance, l.528:
**On another hand, CAMEO[434-126] depicts important negative anomalies of $NH_4^+$, $NO_3^-$, and $SO_4^{2-}$ especially in China (> 4 µg.m−3, equivalent to 60-80% Figure 10, subplots D, I and N).**

-Lines 239-241: It is unclear why dust estimates are being brought up here.
We thank the reviewer for detecting this mistake. This sentence was part of a previous version which should have been also removed.

-Line 252 and Table: The AMoN network is run by the US National Atmospheric Deposition Program (NADP), not the US EPA. Please ensure the text and tables reflect this.
It is right that this is a misunderstanding from our side, AMoN network belong to the NADP which we use for [$NH_{3(g)}$] only. Annual $NO_2$, $NH_4^+$, $NO_3^-$, and $SO_4^{2-}$ are provided by the EPA. It has been clarified in the text and tables.

-Line 261: Beijing
We corrected it.

-Line 264: high concentrations of what?
We clarified it.

-Related to the comment above, use of panel lettering on plots such as Figures 4-6 and more detailed in-text references would aid in understanding these comparisons. I question if all of these figures are necessary for the main body of the manuscript instead of the Supporting Information, as they are not heavily mentioned or discussed within the main text.
Thank you for the recommendation, we decided to move to the SI, the scatterplot figures presenting the evaluation of surface concentrations using ground-based measurements.
We also applied a letter labelling on the relevant Figures and added more references in the text.

-Page 16: figure caption 6 describes European stations but displays information pertaining to North America. Ensure all figure captions are representative.
We corrected it.

-Line 300: delete “also”. Done.
-Paragraph starting on Line 315: “depositions” should be referred to as just “deposition”. Done.
-Line 322: Change “except in” to “aside from”. Done.
-Line 359: Change “relatively” to “relative”. Done.

-Page 20: The calculations presented in the text are not correct. Ex: The difference between CAMEO and CAMEO[585] should be 59%, not 37%. Please ensure all percent changes are calculated correctly throughout the manuscript. Done.
-Page 23: Figure 11 is referenced in text before Fig 10. Please ensure all figures are referenced in the appropriate order. We doubled check.
-Line 488-489: Why is AeroCom Phase III and/or GISS brought up here when it has not been mentioned before this? Citations should be provided if the authors are comparing their findings to other literature.
Indeed, that was missing. We introduced more carefully the AeroCom initiative at the *Model set up* level and cited the authors when comparing to their results:
**Extensive evaluations of the aerosol component of the LMDZ-INCA model have been carried out during the various phases of Aerosol Comparisons between Observations and Models (i.e. AeroCom (Gliss et al., 2021,Bian et al., 2017).**

-Line 497: Why is the sulfate radiative impact not shown for both CAMEO simulations?
The radiative forcing impact of sulfate is shown in table 6 for all "future" simulations (CAMEO[SSPi]) at the row "RF (TOA)". The value is not given for the historical simulation because we defined this simulation as the baseline for RF and AOD.

This has been newly clarified line 550:
**The all-sky direct radiative forcings at the top of the atmosphere (RF TOA) are presented in Table 6 and are calculated as the difference between the future considered CAMEO radiative fluxes and the historical CAMEO fluxes.**

But also line 188:
**In addition to the concentrations of ammonia-related aerosols and gases exploited in this study, the all-sky direct radiative fluxes at the top of the atmosphere and the Aerosol Optical Depth (AOD) of the various aerosol components are calculated online by the atmospheric circulation model. More details on the radiative fluxes computation can be found in Hauglustaine et al. (2014).**

-Line 499: change "slow down" to "slowed down". Done.

-Page 33. The last numbered paragraph (9) of the summary and conclusions represent future directions, and therefore should not be included as a numbered conclusion item. I believe it would be stronger to end the manuscript with a paragraph or two that bring together the full conclusions of the manuscript, incorporating the future directions noted in the (9) paragraph.
A new section named "Future directions : towards N interactions in ESM" have been added and the old paragraph (9) of the previous section has been moved to this new section.

**In this study, the simulations are designed to isolate the impact of emission changes by keeping meteorological conditions fixed at present-day levels during 2090-2100. Climate change is expected to influence atmospheric chemistry through multiple interrelated factors, such as altered mean and extreme precipitation patterns that affect**

**deposition, warming that could shift key chemical reactions, and wind variations that can affect aerosol transport.**
**In a subsequent study, additional simulations will explore the combined impact of both emissions and climate change by incorporating changing meteorological conditions for atmospheric chemistry.**
**Incorporating the nitrogen cycle into Earth System Models is a recent advancement, as highlighted by Davies-Barnard et al., (2022).**
**Developing interactions of nitrogen compounds is complex due to the intricate processes involved, necessitating readiness in coupling atmospheric chemistry and land components.**
**The studies by Pleim et al., 2019, Vira et al., 2019 and Vira et al., (2022) provide a foundational step toward bidirectional ammonia handling, though not yet fully integrated into existing ESMs.**
**Vira et al., (2022) notes that FANv2 does not currently feed back nitrogen losses to the nitrogen cycle in the Community Land Model, leaving fertilizer nitrogen availability to crops unaffected.**
**Our present approach does include feedback from nitrogen loss affecting available soil nitrogen for vegetation, even without a bidirectional scheme yet exploited.**
**Additionally, in the CAMEO framework, we incorporated nitrogen biomass removal for livestock needs, ensuring nitrogen and carbon budget accuracy.**
**Current efforts are focusing on developing nitrogen species exchanges at the atmosphere-surface interface in the IPSL-ESM, aiming to assess chemical and climate impacts through interactive coupling.**