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 1:

The manuscript “Evaluating present-day and future impacts of agricultural ammonia emissions on atmospheric chemistry and climate” by Beaudor et al. examines the impacts of ammonia (NH3) emissions on atmospheric chemistry and climate. The authors used the chemistry-climate model (CCM) LMDZ-INCA to simulate present-day and future atmospheric processing of NH3 (atmospheric concentration, aerosol formation and deposition), while also evaluating the climatic consequences (changes in radiative forcing RF and aerosol optical depth AOD) under different shared socio-economic pathways (SSPs). The novelty of the study lies in the incorporation of a new NH3 emission input from a global land surface model, ORCHIDEE-CAMEO to the CCM. The model results were compared with satellite observations and ground-based measurements, demonstrating good agreement. This study contributes to adding knowledge of future projections of atmospheric chemistry and climate, with an emphasis on the role of NH3 emissions. The manuscript aligns well with the scope of Atmospheric Chemistry and Physics, and I recommend publication after the authors address a few points.

We are thankful for the fruitful comments given by Reviewer 1. Our point-by-point responses can be found below.

General comments:

1. A key improvement the authors could make is to explain why the CCM was used to simulate the period from 2004 to 2014, but the model results were compared with ground-based measurements from 2015. It seems less convincing to compare modelled outputs averaged over 11 years to a different year that is not included in the simulation period. I understand the same-year comparisons might not always be possible because of insufficient data, but this does not appear to be an issue for this study. The emission model (Beaudor et al. 2023 GMD) provides NH3 emissions from 2005 to 2015, and meteorological input from the ERA-interim reanalysis for 2015 should be accessible. Measured datasets of annual surface NH3 concentrations in year 2010 are available as shown in Fig. 4 in Ge et al. (2021 GMD).

This also raises the question of why NH3 concentration comparisons for 2010 were not included in the analysis.
I think it is important for the authors to either:

A) Extend the CCM simulation by an additional year, incorporating the corresponding emission input, so that year 2015 can be included and directly compared with the observations from the same year.
B) Provide a clear justification for comparing model outputs with measurements from different years, along with a discussion of the uncertainties and implications associated with this approach, particularly given that there is inter-annual variability in NH_3 emissions.
Regardless of how the authors choose to address this question, I strongly encourage to include an evaluation for the year 2010, i.e., by adding annual NH3 comparisons for different regions to Fig. 4 to 6.

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 Figures 4 to 6 in the revised manuscript and adjusted Table 4 and text accordingly.
The comparison for 2010 has been added to the Supplementary Material and not in the main text because the results 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 in 2010 compared to 2015, these results are presented in the Supplementary Material (Fig. S7, S8, and S9).

2. It would be helpful to provide more details in the Method section. I find it unclear on the description of how RF and AOD are calculated.
This description has been added in the Methods section, l.215:
Multiple radiative forcings (RFs) and aerosol optical depths (AODs) related to changes in atmospheric composition due to agricultural emissions are calculated online during the LMDZ-INCA simulations.

As also mentioned by Terrenoire et al., 2022, the radiative calculations in the general circulation model (GCM) utilize an enhanced version of the ECMWF scheme established by Fouquart et al., 1980 for the solar spectrum and by Morcrette et al., 1991 for the thermal infrared spectrum.
The short-wave spectrum is segmented into two ranges: 0.25–0.68 and 0.68–4.00 µm. The model incorporates the diurnal variation of solar radiation and permits fractional cloud cover within a grid cell. These RFs are computed as instantaneous, clear-sky, and all-sky forcings at both the surface and the top of the atmosphere. To evaluate the future effects of ammonia emissions on aerosol concentration and climate, the all-sky direct radiative forcings are determined by subtracting the historical CAMEO radiative fluxes from the future simulation being analyzed.
In Section 5.3, the all-sky forcings at the top of the atmosphere and AOD will be discussed for aerosols, similar to what was done by Hauglustaine et al., 2014.

3. It would be interesting to see a full NH3 budget. E.g., I would like to see the authors show how much NH3 contributes to the formation of N aerosols, in addition to what has been reported in Table 5.
We agree with the Reviewer, this information was missing so we added the NH_3 loss from the NH_4 formation in Table 5. Please note that we revised the entire table and numbers might have changed because of an inconsistent total area used for the calculation of the different terms. We also added biomass burning to the total NH_3 emissions which were missing.

Specific comments:
Line 87: The authors can provide a timeseries of the new NH3 emissions that were used for the modelling. This figure can be put to the Supplementary materials, and readers can learn the inter-annual variability in the emissions.
A figure has been added to the SI representing the evolution of the global agricultural NH_3 emissions for SSP2-4.5, SSP4-3.4 and SSP5-8.5 from CAMEO under future climate (SSP2-4.5 is shown but not exploited in the present study).

Line 100: Why estimated agricultural NH3 emissions were reported to be 44 Tg N per year from 2005 to 2015 in the emission paper (Beaudor et al. 2023 GMD), but it turns to 35 Tg N per year for 2004 to 2014 in this study? Why is there such a big difference?
Thanks for raising this question.
The reference paper (Beaudor et al. 2023, GMD) presents CAMEO simulations from a different setup from the CMIP6 framework used in this study and the latest submitted study to JAMES (https://essopenarchive.org/doi/full/10.22541/essoar.170542263.35872590/v1).
In the framework of phase 6 of The Coupled Model Intercomparison Project, a common set of experiments is designed and aims to provide the boundary condition and forcing dataset needed for CMIP6.
Regarding the boundary conditions, for self-consistency, CAMEO for the CMIP6 framework exploits the 3-hourly near-surface meteorological fields simulated by the Institut Pierre Simon Laplace (IPSL) Earth System Model :IPSL-CM6A-LR ESM (Boucher et al., 2020), in the context

of CMIP6 for near-surface air temperature, specific humidity, wind speed, pressure, short- and longwave incoming radiation, rainfall, and snowfall.
The reference paper is based on the Climatic Research Unit (CRU) and Japanese reanalysis (JRA) dataset (CRU-JRA V2.1) (Harris et al., 2014) (preprocessed and adapted by Vladislav Bastrikov, LSCE, July 2020), provided at 6 h time steps.

On another hand, “forcing files” (such as N deposition, N fertilizers etc..) can be less realistic than those used in our reference version, even though they have been extensively used in Land Surface Models and evaluated.
For instance, data for synthetic fertilizer in CMIP6 does not assume that managed grasslands are fertilized.
For this reason, the reference simulation presented in the “emission” paper does not exploit the latest data from CMIP6 as forcing files.
For the present study, to be consistent with the forcing files from CMIP6 for the different SSPs, another historical CAMEO simulation with CMIP6 data was necessary.
As mentioned above, the fertilization budget is much lower in the “CMIP6” simulation (97 TgN/yr against 118 TgN/yr) while the manure produced is 5 TgN/yr less.
The lower total N input in the “CMIP6” simulation explains its lower soil emission budget.
The region where the fertilization use budget is the most reduced in the “CMIP6” simulation is Asia (44% less in India and around 30% in China and Asia Tropical South).
However, synthetic fertilizer use in Africa and Latin America is higher in the “CMIP6” simulation than in the reference simulation (of about 48% and 30%).
Even though fertilizer use is more important in these regions, the resulting regional soil emissions also decrease due to its weak contribution to the total N input and a small reduction in N manure application (-13% and -2%).

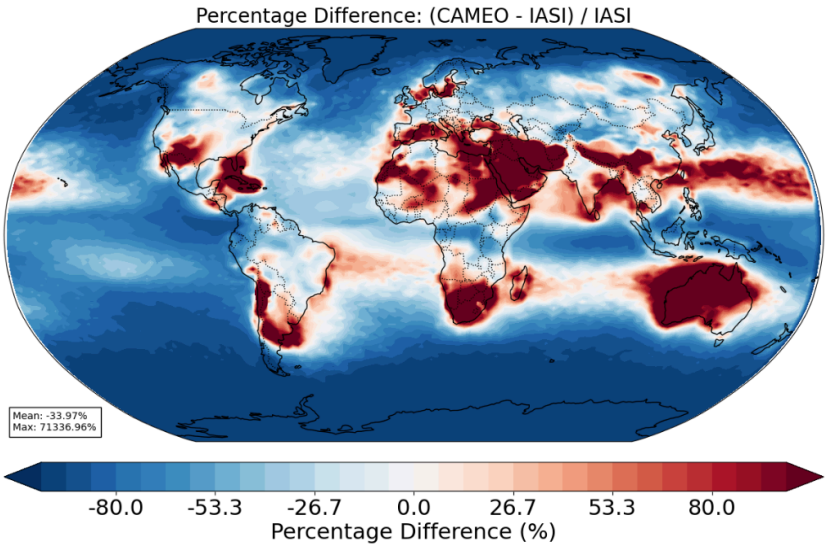
The following paragraph has been added:
Please note that due to a different set of input/boundary conditions data, the agricultural ammonia emissions from the present study are 9 TgN/yr lower than the one reported in the reference study (Beaudor et al., 2023, GMD). This difference is mainly explained by the non-consideration of managed grassland in the CMIP6 synthetic fertilizer forcing which led to a total fertilization input of 97 TgN/yr against 118 TgN/yr in the reference study.
On another hand, the different climatic forcings may also impact the emissions.
For self-consistency, CAMEO for the CMIP6 framework exploits the 3-hourly near-surface meteorological fields simulated by the Institut Pierre Simon Laplace (IPSL) Earth System Model :IPSL-CM6A-LR ESM (Boucher et al., 2020), in the context of CMIP6 for near-surface air temperature, specific humidity, wind speed, pressure, short- and longwave incoming radiation, rainfall, and snowfall.
The reference paper is based on the Climatic Research Unit (CRU) and Japanese reanalysis (JRA) dataset (CRU-JRA V2.1) (Harris et al., 2014) (preprocessed and adapted by Vladislav Bastrikov, LSCE, July 2020), provided at 6 h time steps.

Line 125: Is the emission resampled to fit the resolution of the CCM?
Yes, indeed, the emissions are regridded at the LMDz-INCA spatial resolution and read at a monthly time-step.
The following sentence has been added:
The CAMEO emissions are, first, carefully regridded onto the model grid through a preprocessor program and provided at a monthly time resolution to the chemistry-transport model.

Line 154-161: Which meteorological variables were used for the modelling? What is the spatial and temporal resolution of the meteorological inputs? Another question is since LMDZ-INCA is a CCM, what is the reason for not using the weather fields generated by itself?
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 line 238.

P9: Regarding Fig. 1, please consider showing the percentage difference for map (d) CAMEO – IASI.

As indicated in the following figure, the percentage difference map leads to extremely high values in the non source regions.
In these zones, there is little NH_3 or its presence is not easily detected by IASI.
Therefore this representation of the differences is not optimal.
We rather mentionned percentages in the manuscript for the emissions zones, for instance:
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 224-225: By what evidence can the authors claim that IASI observations does not reveal a "unique peak" which is a modelled feature?

This sentence has been reformulated as follows:

In the US and Europe, the CAMEO columns show a unique peak (0.7 molecules x10^{16}cm^{-2}) during summer, while the IASI observations inform about a lower maximum value (0.5 and 0.4 molecules x10^{16}cm^{-2}, respectively) reached over March-September.

Line 235-236: I see CAMEO shows the peak in the same months as CEDS for India (Fig. 3). Can you check?

Thanks for noticing this inaccuracy. We corrected it as follows:

In India, both CAMEO and CEDS simulate a peak value occurring 2 months earlier than that measured by IASI, but the value is 1.5 times higher with CEDS than with CAMEO.

P11: Figure 3 shows that CEDS performs better in EU than CAMEO.

A more explicit statement is added regarding the better performance of CEDS:

In Europe, CEDS surpasses CAMEO when it comes to the magnitude of seasonal variations.

Line 245: Why not compare annual NH3 concentrations in 2010?

This aspect has been addressed in the general comment responses.

Line326: Why LMDZ-INCA uses a low constant? What is the implication?

We apologize but it is a confusion from our side, the Henry's law constant has been updated according to Bian et al., 2017.

We corrected this aspect in the manuscript.

P20: As suggested, it would be interesting to show the aerosol formation in Table 5.

This aspect has been addressed above.

Line 359-360: I think there is a problem with the calculations. E.g., for CAMEO[585], the increase is (0.27-0.17)/0.17 = 0.59, 59 % rather 37 %. Please do check the numbers in the following text and in the abstract.
Thank you very much for your careful reading, we corrected the numbers.

Line 388: Why H2SO4 is not included for TS in Equation 1?
According to Seinfeld et al., 1997, Metzger et al., 2012 and Hauglustaine et al., 2014 SO_4^{2-} represents several forms that are neutralized by ammonia (more precisely it refers to all of H_2SO_4, NH_4HSO_4, $(NH_4)_3H(SO_4)_2$, and $(NH_4)2SO_4$).
It has been clarified in the manuscript l.487:
(TS, including all forms of SO_4^{2-} as H_2SO_4, NH_4HSO_4, $(NH_4)_3H(SO_4)_2$, and $(NH_4)2SO_4$)

P23: I think it is helpful to explain why look at different pressure levels.
The different pressure levels help to understand the persistence of ammonia in the atmosphere even though it is known as a relatively short-lived species. In addition at the surface we can not distinguish between the different regimes properly because of the excess of NH3.
To be clearer this sentence has been added:
To gain a better understanding of the behavior of ammonia and its persistence in the atmosphere under future scenarios, we have selected different pressure levels, including surface level, 900 hPa, and 500 hPa.

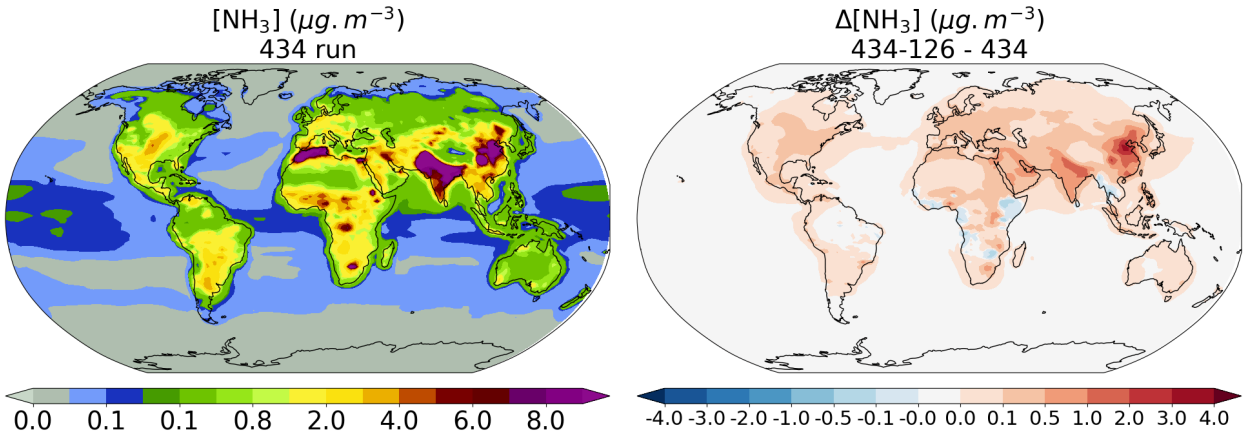
Line 404-405: Please restructure the sentence “It is explained by the reduced amount of NH3 …” to improve the clarity.
We restructured the sentence as:
The decrease in NH_3 can be attributed to its rapid transformation into NH_4 at pressures of 900 hPa and 500 hPa.

Line 416-418: Why is there a shift from nitrate-rich to ammonia-rich at 900 hPa, while the chemical domains at the surface does not change much?
Indeed this was unclear and was in fact a comparison to the historical CAMEO simulation.
We clarified as follows:
In this region, compared to the CAMEO simulation, there is a noticeable expansion of the nitrate-rich and ammonia-rich domains at 900 hPa which is explained by relatively higher NH_3 concentration and a stronger limitation by HNO_3 availability.

Line 425-426: The sentence is not very clear to me. The effect of what?
The impact of both NOx and SO_2 emissions decreases on ammonia abundance. It has been clarified.

Line 426-428: TBH, I barely see the difference between 434-126 and 434 over India, Europe and the US…

Indeed by plotting the absolute difference between the two simulations, it appears that the increase in the ammonia concentration is the highest over China but non negligible over multiple regions (India, the US, Middle East, North Africa and Europe).
The sentence has been modified:
This is also confirmed by comparing NH3 from CAMEO[434-126] and CAMEO[434] where NH3 emissions are identical but a slightly stronger impact on the concentrations is highlighted for instance in China and India.



Line 440: What does “levels” mean here? Emissions?
Yes, it has been clarified.

P25: I feel there is a need for better referring to figures in this whole section. Sometimes it is difficult to follow the text without looking at the figures, but there is no clear referring.
We added more references to the figures when needed throughout this specific section.

Line 448: Delete “Finally,”.
It has been done.

Line 458: CAMEO[434-370] or CAMEO[585]? Please check Fig. 13.
It is actually for the 3 simulations a similar pattern. We corrected it accordingly.

Line 462-463: Why attribute the same NOy deposition between CAMEO and CAMEO[434] to identical NOx emissions? Figure 10 shows that there are higher [NO3 -] over EA.
Indeed, we agree that this is an uncomplete interpretation of our result.
We analyzed it differently:
There are minimal spatial differences in the deposition of NOy between CAMEO and CAMEO[434] (and CAMEO[585]), as constant NOx emissions lead to a balancing effect, resulting in decreased HNO3 deposition and increased NO3- deposition, especially in China.

Line 472-474: This is a long sentence. I suggest split into short sentences to improve readability.

We adopted this recommendation.

Line 478-479: Why nitrate AOD is mostly increase in CAMEO[434-126]?
Emissions of SO_2 are much lower in that scenario than in the other ones.
As NH_3 is reacting with the sulfate in priority to form ammonium sulfate aerosols, there is much more available NH_3 to form ammonium nitrate aerosols. This counterbalances largely the NO_x emission reductions.
This explanation has been incorporated in the main text.

Line 503: How is the value 1.6 Tg(N2O)yr-1 calculated? Same question for the value of 2.9 in line 508.
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 non-negligible source 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**

The factor used to convert Tg(N2O)/yr into TgN/yr is 1.58.

We apologize for this mistake, the 2.9 Tg(N2O)/yr value is not the correct one for SSP434-370 (this one corresponds to the production under SSP434), we corrected the value in the text to 3.7 Tg(N2O)/yr (the percentage given was correct).

P1 in SM: I think the authors should consider using maps that show the differences between emissions from future and present-day.
We agree on that suggestion and changed Figure S1 for anomalies instead.