The authors would like to thank the reviewers for their thoughtful comments. Responses from the authors are below in red (original reviewer comments are in black).

Reviewer #1

First, the bidirectional flux scheme implemented in this work primarily represents ammonia exchange between the atmosphere and soil or ground surfaces, which is most relevant for agricultural soils and fertilizer-related emissions. In the inversion, however, satellite NH3 retrievals appear to be assimilated over all land areas without screening by land-use type. Satellite observations measure the total atmospheric NH3 column, which integrates signals from all nearby sources. If the inversion does not explicitly separate or mask different land-use categories, there is a risk that ammonia originating from non-soil sources, such as livestock facilities or urban emissions, may be incorrectly attributed to soil-related parameters in the bidirectional scheme. In such regions, the retrieved atmospheric ammonia may reflect a combination of emission processes that are not fully represented by the soil-based bidirectional scheme.

In the submitted draft of the manuscript, this issue was touched upon in lines 244-248, but I can now see that the point made in these lines was not sufficiently explained and needs further clarification (which will be added to the next draft version of the paper).

Lines 244-248 of the (first) draft of the manuscript contained the text:

"Γp describes the sources of ammonium to the ground and can have contributions from, for example, fertilizer application or waste from livestock. … In this work, Γp will represent agricultural sources (fertilizer/livestock), while separate non-agricultural ammonia emissions that account for $\lesssim$ 1% of total emissions will be accounted for using the unidirectional emissions model."

So the bidirectional flux model is to represent emissions from all agricultural source, which in this case are from both fertilizers and livestock, that account for ~99% of the total ammonia emissions. However, it was not explained why we treat the livestock emissions with the same model as the fertilizer emissions.

The assumption being made here is that livestock ammonia emissions primarily originate from excreted nitrogen as manure. This assumption is made for ammonia livestock emissions in CMAQ and GEOS-Chem (see Zhu et al. (2015a) and Zhu et al. (2015b)). The review article by Lee et al. (2025) examines ammonia emissions from beef cattle and concludes that most of the ammonia emissions from beef cattle are from manure (mostly from urine). This assumption is made for all livestock types, and so the livestock emissions can be thought of as a type of fertilizer emissions.

The diurnal pattern of the livestock ammonia emissions is given by Eqs. (1) to (3) of Zhu et al. (2015a) and by Eq. (1) in Zhu et al. (2015b). These equations are essentially Eqs. (2) and (5) of this manuscript, except written in the form of an average or total emissions term modulated by a time-dependent modulation term. In Zhu et al. (2015a) and Zhu et al. (2015b), there isn't an explicit dependence of the manure pH, but in these works the livestock emissions are being modeled in the same manner as the fertilizer emissions.

In this manuscript, the ammonia source terms are label as either ‘ground’ or ‘soil’ but this was meant to be inclusive of manure from livestock. However, this was not made sufficiently clear in the first version of the paper and so the next version of the manuscript will have text added to clarify this point.

The paragraph in the manuscript that was quoted above has been changed to:

“Γp, which describes the sources of ammonium to the ground, is taken as a spatially-varying 2D field on the same horizontal grid as described in Section 2, allowing for a unique field for every month. In this work, Γp will represent agricultural sources (both fertilizer and livestock), while separate non-agricultural ammonia emissions that account for $\lesssim$ 1% of total emissions will be accounted for using the unidirectional emissions model. As ammonia emissions from cattle originate primarily from excreted nitrogen in manure (Lee et al., 2025), we make the same assumption as in Zhu et al. (2015b) in which all ammonia emissions from livestock are assumed to originate from manure. We can then treat both the livestock and fertilizer emissions using the same emissions modeling framework. As such, model parameters that reference the ground or soil should be understood to be inclusive of manure produced by livestock.”

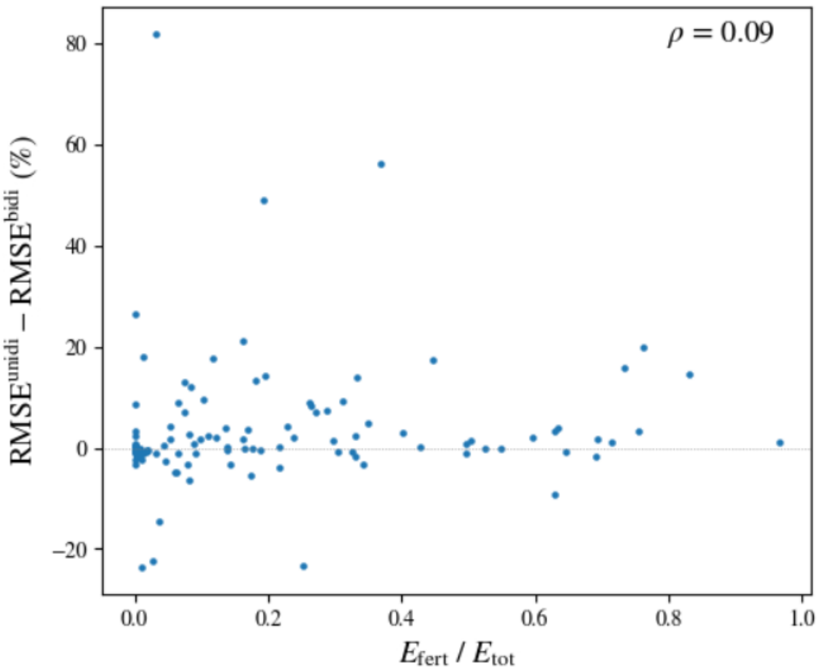
Zhu, L., Henze, D., Bash, J., Jeong, G. R., Cady-Pereira, K., Shephard, M., Luo, M., Paulot, F. & Capps, S. (2015a). Global evaluation of ammonia bidirectional exchange and livestock diurnal variation schemes. Atmospheric Chemistry and Physics, 15(22), 12823-12843.

Zhu, L., Henze, D. K., Bash, J. O., Cady-Pereira, K. E., Shephard, M. W., Luo, M., & Capps, S. L. (2015b). Sources and impacts of atmospheric NH3: Current understanding and frontiers for modeling, measurements, and remote sensing in North America. Current Pollution Reports, 1(2), 95-116.

Lee, M., Auvermann, B. W., Tedeschi, L. O., Koziel, J. A., Brandani, C. B., Gouvêa, V. N., Smith, J. K. & Casey, K. D. (2025). Ammonia emissions from beef cattle feedyards: a review. Frontiers in Animal Science, 6, 1608387.

This could also potentially explain why the model performance improvements appear strongest in cropland-dominated agricultural regions, where soil-related emissions are more consistent with the assumptions of the bidirectional model.

I tried to check if the bidirectional flux model’s performance is in fact better in cropland regions. I made a scatter plot with the data from Fig. 11 that plotted ‘$RMSE^{unidi} – RMSE^{bidi}$‘ on a map, but instead plotted ‘$RMSE^{unidi} – RMSE^{bidi}$‘ on the y-axis and the ratio of the fertilizer NH_3 emissions E_{fert} to total NH_3 emissions E_{tot} for each point on the x-axis, where E_{fert} and E_{tot} are taken from the emissions inventory (plot shown below). The correlation coefficient for this scatterplot is in the upper right corner. The value of the correlation coefficient is low and I can’t see much of a relationship in the scatterplot. So I'm not sure if the model does perform better in these regions as compared to regions where the livestock emissions dominate.



I therefore suggest that the authors discuss whether any land-use filtering or weighting was applied when assimilating satellite observations, how non-soil ammonia sources may influence the inversion results, and whether the optimized parameters could be biased by the presence of mixed emission sources.

Because the bidirectional flux model was used both fertilizer and livestock emissions and urban emissions only represents $\lesssim$ 1% of total emissions, no land-use filtering was done.

Second, the temporal resolution of the simulations and inversions is not entirely clear from the manuscript. The paper mentions that a month's worth of satellite retrievals is used in each inversion, resulting in monthly mean parameters. However, it would be helpful if the authors could clearly state the temporal resolution of the model simulations, for example whether transport and chemistry are simulated at hourly time steps, the temporal aggregation used in the inversion procedure, and whether the satellite observations are assimilated at their native temporal resolution or aggregated before assimilation. Given the strongly episodic nature of ammonia emissions, particularly fertilizer-induced emission pulses following application events, the temporal resolution could influence the ability of the inversion system to properly constrain bidirectional exchange parameters.

In Section 2 that describes the GEM-MACH model, the following line has been added:

"Meteorological/physics variables are integrated using a time step of 5 minutes, whereas the chemistry variables use a time step of 15 minutes, and model output is saved at every 15 minutes."

At the end of Section 7.1 ('Inversion Procedure'), this line has been added:

"Retrievals are compared to the GEM-MACH model output (available at 15 minute increments) closest to the retrieval time."

At the end of Section 7.2 ('Inversion Parametrization'), this line has been added:

"The ensemble GEM-MACH output was saved only at the top of the hour, in contrast to the unperturbed GEM-MACH runs that had output saved at every 15 minute time step, to reduce the storage requirements of the ensemble."

There was no aggregation of the retrievals prior to input into the inversion system.

Third, the model evaluation focuses primarily on surface ammonia concentration observations from the AMoN and NAPS monitoring networks. While these datasets are valuable for evaluating atmospheric ammonia concentrations, ammonia deposition observations are also widely used to constrain ammonia emissions and nitrogen budgets. Because bidirectional exchange influences both emissions and deposition processes, deposition measurements, such as ammonium wet deposition observations, could provide an additional and independent constraint on model performance. It would therefore be useful for the authors to discuss whether deposition observations could be used in future work to further evaluate the optimized bidirectional scheme.

This line has been added to the end of the conclusions section:

"Future evaluations could extend these validations by including comparisons to ammonium wet deposition observations to provide an additional validation data set."

Line 110: Please clarify the version and type of the "2011-based projected 2017 inventory for the United States." The 2011 US NEI inventory has undergone several updates, and it would be helpful to specify whether the inventory used here is the emission factor–based inventory or one generated using a bidirectional scheme.

This has been changed to:

"This set of emissions were generated using a 2013 emissions inventory for Canada (https://www.canada.ca/en/environment-climate-change/services/pollutants/air-emissions-inventory-overview.html, last access: 14 March 2025), a 2011-based projected 2017 inventory (version 6.3) for the United States (https://www.epa.gov/air-emissions-modeling/2011-version-63-platform, last access: 24 February 2025) from the US Environmental Protection Agency (EPA), and a 2008 inventory (version 6.2) for Mexico (https://www.epa.gov/air-emissions-modeling/2011-version-62-platform, last access: 24115 February 2025) from the US EPA. The emissions inventories from the EPA were derived using a traditional emission factor‑based method instead of the bidirectional flux-based inventories used in later versions of the National Emissions Inventory (NEI)."

Line 175: "nonzero" should be written as "non-zero."

Changed.

It would also be helpful if the authors included a discussion of uncertainties associated with the inversion results. A brief discussion of how uncertainties in satellite retrievals, prior emissions, and model parameter assumptions may influence the inversion results and the retrieved parameters would help readers better interpret the robustness of the conclusions.

A new section (Section 8.5) has been added to address this comment, as well as a comment made by Reviewer #2. The first paragraph of this section is:

"	We conclude with a brief examination of the relative sensitivity of the inversions to the inversion model parameters. While there is a noticeable reduction in the a posterior uncertainties for both Γp and pHg, the reduction in uncertainties for Γp was much larger than that for pHg in many regions (see Figure S5 of the Supplement). Furthermore, the number of degrees of freedom for the inversions (Rodgers, 2000) associated with Γp are 2 to 8 times larger than that associated with pHg. Overall, the inversions are better able to constrain Γp then pHg ."

This new text references a new figure added to the Supplement (Fig. S5), which is included below.

Rodgers, C. D.: Inverse methods for atmospheric sounding: theory and practice, vol. 2, World scientific, 2000.

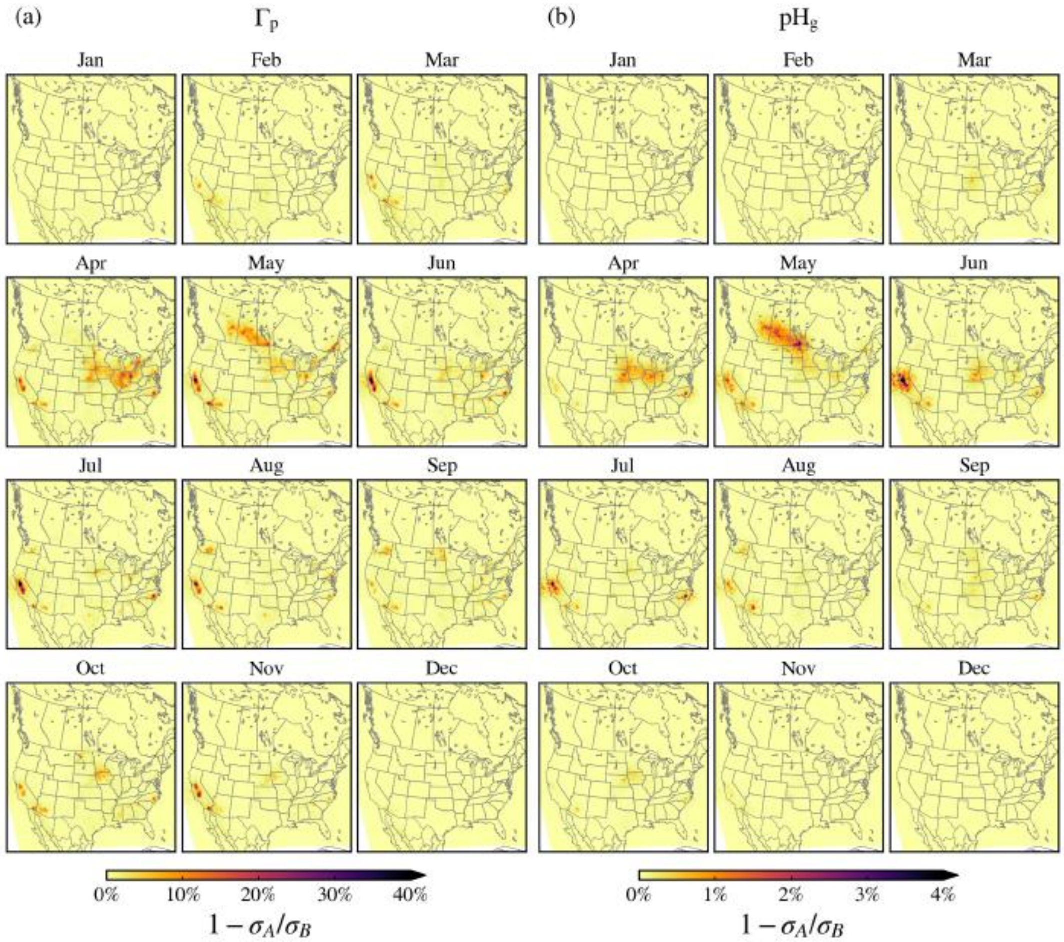


Figure S5. Reduction of the analysis (a posterior) uncertainties σ_A relative to the background (a priori) uncertainties σ_B for the inversion parameters (a) Γ_p and (b) pH_g.

<u>Reviewer #2</u>

Specifically, the authors chose to optimize the ammonium source term Γp that "sets the levels of ground ammonium" and the soil pH which "determines the rate at which the ammonium pool reaches equilibrium with ammonia in the atmosphere". This choice might need some more justification and discussion because physically the two terms are related/correlated, with H+ being part of Eq (13) that calculates Γp, which will make the inversion more complex and make it tricker to interpret the results.

The intention of displaying Eq. (13) was to show that the model parameters used in the bidirectional flux model, Γ_p and τ_p, could be written in terms of individual source terms, even though specifying these individual pathways is not necessarily for this model. However, I think Eq. (13) as written in the previously submitted draft has given the impression that Γ_p, τ_p , and pH are not independent model parameters, but this is not the case.

In the first draft of the manuscript, Eq. (13) and the sentence that follows is

"

$$\Gamma_{\mathrm{p}} = \frac{\tau_{\mathrm{p}}}{h[\mathrm{H}^+]} \sum_{j=1}^{\mathrm{N_p}} F_{\mathrm{p}}^{(j)} \quad (13a)$$

$$\tau_{\mathrm{p}}^{-1} = \sum_{j=1}^{\mathrm{N_p}} (\tau_{\mathrm{p}}^{(j)})^{-1} \quad (13b)$$

where $F_{\mathrm{p}}^{(j)} = \chi_{\mathrm{p}}^{(j)}/R_{\mathrm{p}}^{(j)}$ is the ammonium flux from pathway j and $\tau_{\mathrm{p}}^{(j)} = R_{\mathrm{p}}^{(j)} C$ is its associated time scale. "

As Eq. (7) is

$$C = h\frac{T_{\mathrm{g}}}{A} e^{B/T_{\mathrm{g}}}[\mathrm{H}^+] \quad (7)$$

so then τ_{p} in Eq. (13b) can be rewritten as

$$\tau_{\mathrm{p}}^{-1} = \sum_{j=1}^{\mathrm{N_p}} (R_{\mathrm{p}}^{(j)} h \frac{T_{\mathrm{g}}}{A} e^{B/T_{\mathrm{g}}}[\mathrm{H}^+])^{-1}$$

Substituting the expression above for τ_{p} into Eq. (13a) gives

$$\Gamma_{\mathrm{p}} = \frac{1}{h[\mathrm{H}^+]} \left[\sum_{j=1}^{\mathrm{N_p}} \left(R_{\mathrm{p}}^{(j)} h \frac{T_{\mathrm{g}}}{A} e^{\frac{B}{T_{\mathrm{g}}}}[\mathrm{H}^+] \right)^{-1} \right]^{-1} \sum_{j=1}^{\mathrm{N_p}} F_{\mathrm{p}}^{(j)}$$

$$= \frac{T_{\mathrm{g}}}{A} e^{\frac{B}{T_{\mathrm{g}}}} \left[\sum_{j=1}^{\mathrm{N_p}} \left(R_{\mathrm{p}}^{(j)} \right)^{-1} \right]^{-1} \sum_{j=1}^{\mathrm{N_p}} F_{\mathrm{p}}^{(j)}$$

so that the factor $[\mathrm{H}^+]$ in the square brackets cancels out the factor of $[\mathrm{H}^+]$ out front.

According to Eq (13), Γp would monotonically increase with soil pH (hopefully I didn't get it the other way around).

So Γ_{p} is independent of $[\mathrm{H}^+]$ (and pH) because there is a factor of $[\mathrm{H}^+]$ 'hidden' in τ_{p} that cancels out the $[\mathrm{H}^+]$ in the denominator of Eq. (13a). But I can see how this was not very clear from how Eq. (13) was presented in the first version of the manuscript. In the next version of the manuscript, I've changed Eq. (13a) to

$$\Gamma_{\mathrm{p}} = \frac{T_{\mathrm{g}}}{A} e^{\frac{B}{T_{\mathrm{g}}}} \sum_{j=1}^{\mathrm{N_p}} \chi_{\mathrm{p}}^{(j)}/R_{\mathrm{p}}^{(j)} \left[\sum_{j=1}^{\mathrm{N_p}} 1/R_{\mathrm{p}}^{(j)} \right]^{-1}$$

which can be found from simply substituting $F_{\mathrm{p}}^{(j)} = \chi_{\mathrm{p}}^{(j)}/R_{\mathrm{p}}^{(j)}$ into the equation above. I think this form is clearer since it directly references the variables that are labeled in Fig. 2 (i.e. the $\chi_{\mathrm{p}}^{(j)}$ and $R_{\mathrm{p}}^{(j)}$) and has a straight-forward interpretation, in that Γ_{p} is a weighted sum of the ammonium source terms $\chi_{\mathrm{p}}^{(j)}$ weighted by the conductivity $1/R_{\mathrm{p}}^{(j)}$ associated with each pathway, with a factor of $\frac{T_{\mathrm{g}}}{A} e^{\frac{B}{T_{\mathrm{g}}}}$ to transform from a concentration to a (unitless) potential term.

Mathematically the background error covariance matrix for the parameters β would need to account for such correlation with off-diagonal non-zeros to handle that correlation. In interpretation of the results, one has to also keep in mind that the two are related. Therefore, higher soil pH or higher Γp will have the same effect in increasing ammonia fluxes and concentrations. It then becomes interesting how CrIS can differentiate the two. This is an important question about the method, so I suggest the authors expand their discussions reasoning on such a choice if they had considerations not yet discussed in the paper.

Hopefully changing how Eq. (13a) was presented clears this up.

Looking at the results (Figure 6), I can see vast regions where both of the two have positive increments and also places where they go opposite directions (e.g., Alberta in October). This might be some interesting results to discuss further.

The pH increments are always positive because decreasing the pH from its background value has practically no effect on the system, since that would increase τ_{a} from a few months to years, but τ_{a} much more than a month is already long enough that the ammonium pool never really equilibrates with the atmospheric ammonia and only really changes with changes to the source ammonium (Γ_{p}).

I also wonder why the authors chose to start with soil pH=5 everywhere instead of some global dataset soil pH?

The reason for this choice was explained in Section 7.2, but it might not have been sufficiently clear. The a priori used was the assumption that the emissions potentials were time-independent, which is the assumption made in many bidirectional flux models, which, for example, was done in Nemitz et al. (2001), Zhang et al. (2010), Wichink Kruit et al. (2012), Whaley et al. (2018), and Davis et al. (2025). The assumption of a low pH value (in this case 5) is equivalent to a time-independent emissions potential. This seemed appropriate as the assumption of a time-independent emissions potential was used in previous ammonia bidirectional work using GEM-MACH (Whaley et al. (2018) and Davis et al. (2025)).

Zhu et al. (2015a) (citation above) do take their soil pH values from the ISRIC – World Soil Information database. I considered doing the same for this work, but in the database I could not sufficiently determine the land-use type for each sample in the database, and so was uncertain about how representative each sample was to the ammonia sources examined in this work and so decided to use the time-independent emissions potential a priori.

I've rewritten part of Section 7.2 to try to make this cleaner, which now read as:

" The background (a priori) values for Γp were set to the values that yield the inventory-derived monthly mean emissions described in Section 3. The background error covariance used for Γp was nearly identical to that described in Sitwell et al. (2022), in which background error standard deviations were set to 50% of the background values (with a minimum standard deviation corresponding to emissions of 0.26 kg ha−1 month−1 to ensure all locations have a non-negligible deviation) and homogenous and isotropic correlations with a half-width at half-maximum of 40 km.

The background values for pHg were chosen so that the resulting values for τ a were much longer than a month. This makes our a priori equivalent to assuming that the emissions potentials are time-independent. This choice was made in part due to time-independent emissions potentials being used in previous studies of ammonia bidirectional fluxes in GEM-MACH (Whaley et al., 2018; Davis et al., 2025) as well as with other models (Nemitz et al., 2001; Zhang et al., 2010; Wichink Kruit et al., 2012). As a pH value of 5 results in τ a values much longer than a month, a uniform background value of 5 was set for pHg (we also note that the mode of the distribution of soil pH values from the World Soil Information Service (WoSIS) (Batjes et al., 2020) is near 5, see Figure S1 of the Supplement). The background error covariance for pHg was constructed in a similar manner to that for Γp, but with a standard deviation of 3. Additionally, a minimum value of 1.5 and maximum value of 8.5 was imposed on the pHg distribution to limit the number of pH values far outside of the range of pH values found in the WoSIS database (see Figure S1 of the Supplement). While this distribution results in more pH values below 5 as compared to the WoSIS database, as all pH values below 5 yield very large values for τ a, these values should only be interpreted as indicating a static emissions potential (and not necessarily to be interpreted as physical values)."

If possible it would be interesting and useful to see inversions just to optimize the fluxes with fixed pH field and how similar/different the results are.

The new section referenced above (Section 8.5) includes the paragraph:

" While the ammonia retrievals from CrIS do constrain Γp more then pHg, including pHg as an inversion parameter can nonetheless have significant effects on the atmospheric ammonia concentration. Inversions were repeated with $\beta = \{\Gamma p\}$ as the sole inversion field, with the values of pHg set to values from the WoSIS pH database. The root mean square differences in the ammonia surface concentrations between inversions with $\beta = \{\Gamma p\}$ compared to inversions with $\beta = \{\Gamma p, pHg\}$ (shown in Figure S6 of the Supplement) can be as large as 10 ppbv in some regions, such as the midwestern US and California's Central Valley, which is comparable to the differences between the unidirectional and bidirectional models (as seen in Fig. 10), although with not as large of a horizontal extent. Including pHg as an inversion parameter also decreases the biases with AMoN observations in May, June, September, and October, but increases the bias in July and August (see Figure S7 of the Supplement)."

New figures (Figs. S6 and S7, included below) have been added to the Supplement to show the effect of keeping the pH values fixed on the atmospheric ammonia concentrations and on the biases with the AMoN observations.

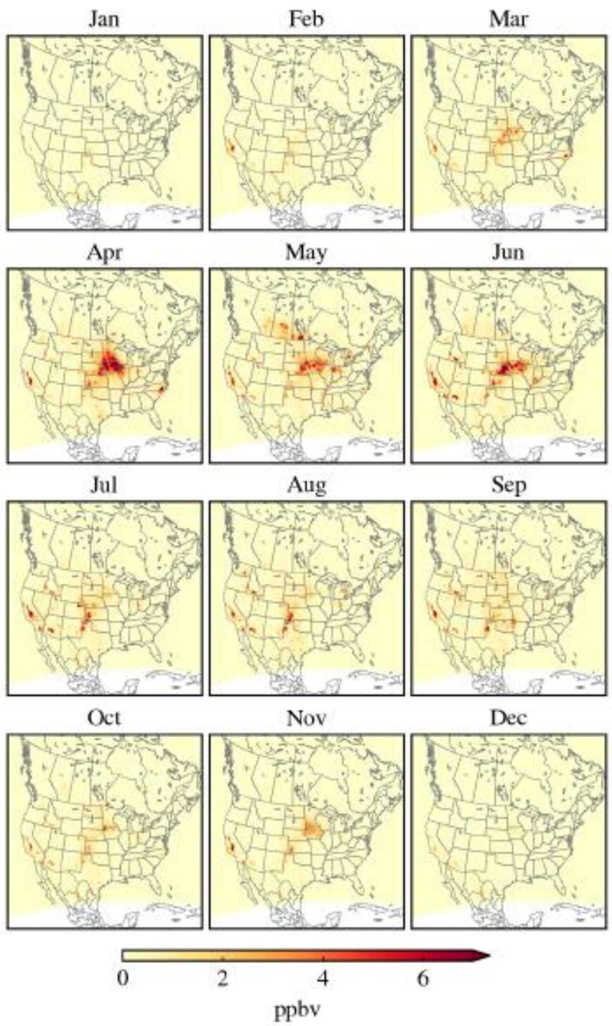


Figure S6. Root mean square differences in ammonia surface concentrations between bidirectional flux models. The first bidirectional flux model has both Γ_p and pH_g derived from inversions. The second bidirectional flux model has only Γ_p inversion-derived and pH_g set from values from the WoSIS pH database.

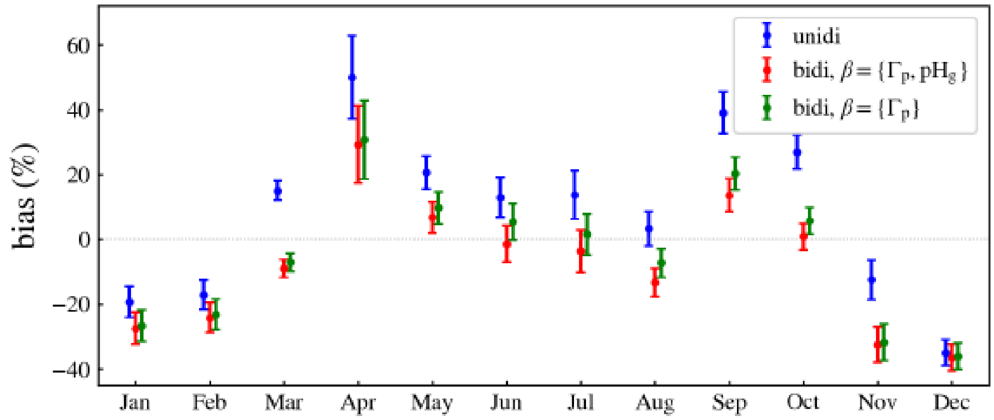


Figure S7. Biases of the unidirectional model (with inversion-derived emissions; blue), the bidirectional flux model with both Γ_p and pH_g set from inversions (red), and the bidirectional flux model with only Γ_p set from inversions and pH_g set from values from the WoSIS pH database (green) as compared to AMoN observations for 2016. Error bars indicate the 1σ confidence interval.