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

Reviewer #2

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

“

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

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

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

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

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 Γ p were set to the values that yield the inventory-derived monthly mean emissions described in Section 3. The background error covariance used for Γ p 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 Γ p, 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.

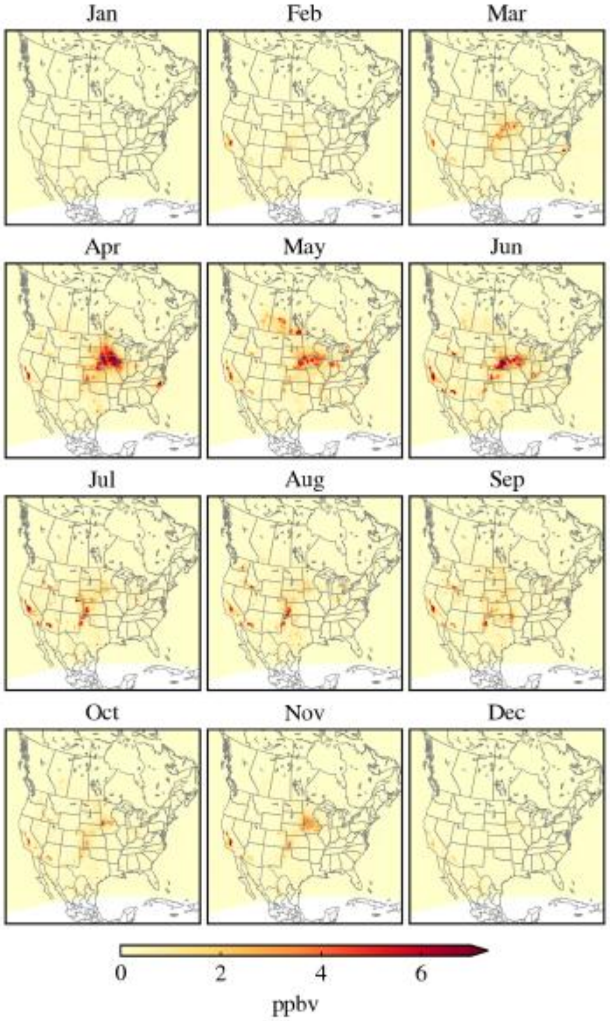

Figure S6. Root mean square differences in ammonia surface concentrations between bidirectional flux models. The first bidirectional flux model has both Γ_p and pH_g derived from inversions. The second bidirectional flux model has only Γ_p inversion-derived and pH_g set from values from the WoSIS pH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

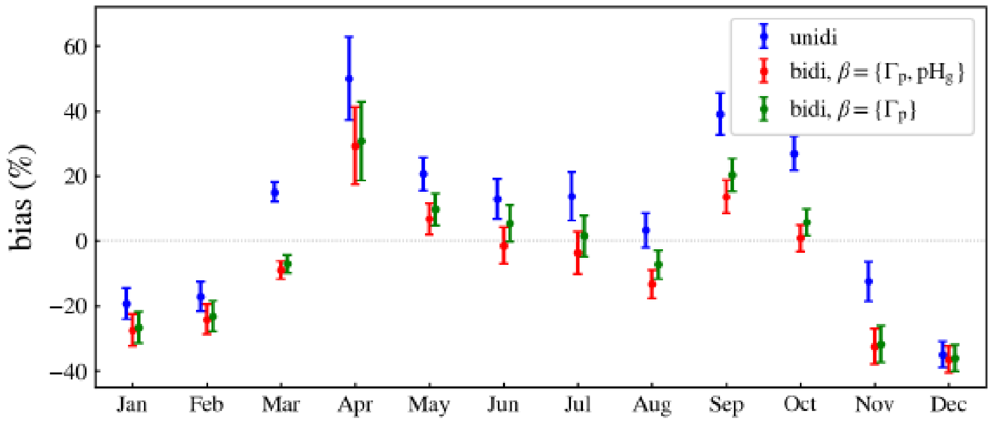

Figure S7. Biases of the unidirectional model (with inversion-derived emissions; blue), the bidirectional flux model with both Γ_p and pH_g set from inversions (red), and the bidirectional flux model with only Γ_p set from inversions and pH_g set from values from the WoSIS pH database (green) as compared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