We thank both reviewers for their comments and suggestions. Responses with text edits where relevant are in blue.

Reviewer 1:

General Description and Recommendation:

The authors use the GEOS-Chem model to characterize reactive nitrogen (NOy) throughout the global troposphere. The characterization follows broad qualitative comparison of the model to NASA DC8 ATom aircraft observations over the remote ocean. The characterization provides details of the budget of prominent NOy compounds, a detailed depiction and overview of the magnitude of reaction pathways forming and destroying NOy compounds, and of emissions and deposition pathways. Some quantification of lifetimes of NOx and NOy against chemical sinks and deposition sinks is also given. The study is an incredibly valuable and much needed state of knowledge of NOy chemistry as represented by a model that keeps pace with the science. The paper is overall well written, the results are clearly depicted in figures throughout the manuscript, and the content is logically presented.

Included below are general concerns or requests for clarity that can be easily addressed by the authors, along with additional specific suggestions to further increase accessibility of the content to ACP readers.

General Comments:

[1] The model is assessed against ATom observations and determined to be robust. The comparison is for particular conditions sampled. Understandably, screening of the ATom observations removes instances like plumes that the grid resolution of GEOS-Chem can't reproduce. But, there are other screening steps applied to remove nighttime observations that don't fit the PSS assumption and to remove data collected over land. Does this then impact the ability to use the model to characterize tropospheric NOy? How consistent are the values in Figures 3-5 to values obtained from the model under the same conditions used to compare the model to ATom?

Our goal in this study is to characterize the NO_y budget as simulated in the current generation of the GEOS-Chem model. However, we certainly do not claim that this is a "validated" model budget. As stated in the manuscript, the comparison of the model with ATom observations (Section 3.1) is intended to serve as a benchmark for the global budget rather than as a validation of the budget. As the reviewer rightly points out, these comparisons are by definition "for particular conditions sampled", and biases are likely to vary with conditions (such as diel variation, location, etc.). Regarding the specific question raised by the reviewer, we cannot assess the consistency between the ATom values and

Figures 3-5 because the reported annual values in Figures 3-5 are derived from monthly mean diagnostics, due to computational constraints. A true validation or evaluation of the model NO_y against a suite of global observations under a range of conditions would indeed likely provide some process-based insight into model bias. This is not part of the design of this study but would be a valuable future contribution. We note that these ideas are acknowledged in our concluding paragraph:
"Future exploration of the NO_y budget on diurnal and seasonal timescales is needed. Additionally, while the focus of this study was to examine NO_y on a global scale, we acknowledge that the relative importance of different NO_y interconversion pathways could vary considerably at the regional scale."

[2] The ATom comparison is mostly a qualitative description of the model performance. The assessment would benefit from providing a sense of scale of biases where the model and ATom observations disagree. This might merely require stating the model normalized mean bias in portions of the manuscript where biases are already identified.
We have added bias and correlation statistics to Figures S1 and S2, and we have now noted these statistics in the text where relevant.

[3] The assessment of the model throughout the results sections is missing reference to pertinent historical (Emmons et al., 1997; Bradshaw et al., 1998; Jaegle et al., 1998) and recent (Lee et al., 2025; Wei et al., 2025) literature that used ATom or predecessor NASA aircraft campaigns along with GEOS-Chem or similar state-of-knowledge models to characterize NOy in particular layers of the atmosphere or to characterize particular NOy compounds throughout the troposphere.
We have added these references.

[4] Figure 3 is an incredibly valuable visualization of the representation of complex tropospheric NOy chemistry represented in a model like GEOS-Chem. The figure could be further enhanced by making it clearer in the caption that units are "Tg N" unless otherwise stated. The first sentence of the caption gives the impression that units throughout are "Gg N". Another change could be to make clearer which pathway corresponds to thermal decomposition of PNs to NO2, as currently the pathway from PNs to NO2 is only labelled as photolysis. The NO to/from HONO seems to not follow the convention used for other reversible processes: a single reversible blue arrow and a black net arrow. Provide a supporting reference for the path from AERNIT (aerosol-phase organic nitrate) to gas-phase HNO3 via hydrolysis, as this seems on first glance like an unfavorable reaction, given it yields a sticky gas-phase compound that prefers to be in the aerosol phase.

Thank you for the suggestions! We have added the following text (underlined below) to the caption to clarify that fluxes are in Tg N yr^{-1}:

> "Modeled NO_y budget for 2016 showing key species and families (dark blue circles) in Gg N and chemical and physical fluxes (arrows) in Tg N yr^{-1}."

We have made changes to the representation of NO_2-PNs and HNO_4-NO_2 chemistry in Figure 3 so that the bidirectional arrow refers only to thermal formation / decomposition and added a label (T) to indicate the role of temperature. We have used a convention of treating photolysis as a distinct process (as seen in other fluxes) from association / recombination reactions, which are represented by bidirectional arrows, and therefore left the representation of HONO chemistry unchanged.

We have renamed AERNIT to orgNITA to mirror the GEOS-Chem naming convention for for individual aerosol organic nitrates (IONITA, MONITA, etc.) and retained orgNIT as the label for gas-phase organic nitrates. This change is also reflected in Figures 3, 6, and 7, in Text S3 and S4, and in Table S2 and S3.

We have added the model's reference for the aerosol organic nitrate hydrolysis pathway (Fisher et al., 2016) to the discussion of organic nitrate chemistry in Section 3.3.

[5] PPN turns out to be a prominent PNs compound, as indicated from the model budget values, but the model comparison to ATom is only given as a figure in the supplementary and the results from this figure are not mentioned in the main manuscript. Even though only 2 campaigns measured PPN, it would be useful to mention how the model compares to the observations in the main manuscript. Then to also say briefly whether the version of the model used in the study includes PPN photolysis, as Wei et al. (2025) suggested that, for the upper troposphere, the lack of PPN photolysis in the model causes a high bias in PPN and low bias in NO2. Horner et al. (2024) addressed this by adding PPN photolysis to the model. PPN photolysis was introduced in a later version 14.5.0 of the GEOS-Chem model, so we have not included diagnostics for this process. However, we have added a reference to the results of Horner et al. (2024) and Wei et al. (2025) as follows:

> "Recent studies (Horner et al., 2024; Wei et al., 2025) note a high bias in the model's representation of PPN, especially in the upper troposphere, and recommend addressing this model bias with the inclusion of PPN photolysis, thereby decreasing PPN and increasing NO_2. Figure S2 in the Supporting Information confirms this high model bias is present in this GEOS-Chem version prior to the implementation of PPN photolysis (131% < NMB < 451% in the Northern Hemisphere and 44% < NMB < 266% in the Southern Hemisphere)."

[6] Provide consolidated definitions of AERNIT, pNO3, NIT, and NITs early in the main manuscript, as it's confusing to follow what these represent. The definitions are in the supplementary, but for ease of following Figures 3-5 and accompanying text, these should also be stated in the main manuscript. Consider renaming AERNIT to orgNIT or NITorg. As written and given the pattern of naming other compounds in the manuscript, AERNIT suggests the sum of all aerosol phase nitrate, rather than the sum of all aerosol phase organic nitrates.
We have expanded the list of definitions at the beginning of Section 3.2 to include all families, and we have renamed AERNIT to orgNITA, as noted in the response to comment #4.

[7] Minor technical issue that appears in multiple locations: parentheses before citations are missing a space between the two (e.g., p. 2, line 45; p. 5, line 133).
Fixed.

[8] Replace all "em dash" instances with periods for ease of comprehension (e.g., p. 3 line 92) and especially p. 13, line 303 (initial interpretation is that em dash is shorthand for "to", indicating a range of values).
Fixed for the majority of cases; retained when intent is clear.

[9] There are a few instances where compounds are defined multiple times throughout the manuscript. HONO and HNO4, for example, are both initially defined on p. 2, then redefined further down p. 2 and HNO4 is again defined on p. 5. Consider removing these redundancies.
We have removed most redundant definitions, retaining only those that improve readability.

Specific Comments:

p.1, line 21-22: Reword "at 500 hPa is 0.10" to "is 0.10 at 500 hPa".
We have changed the sentence to read:

> "The global mean NO_x:NO_y ratio is 0.23 at the surface (over continents it is 0.34) and is 0.10 at 500 hPa."

p.2, line 46 "biomass burning" is referred to as natural, but biomass burning is a combination of wildfires and intentional burning by humans for agricultural practices.
We have replaced the word "natural" with "additional" in that sentence to avoid mis-classification.

p.3, line 88: Delete "a" in "with a several studies noting".
Fixed.

p.3, line 97: Should NOx as precursor of PM2.5 also be acknowledged in this sentence about the motivation for regulating NOx sources?

We have changed the sentence to read:

> "Due to the importance of NO_x as a pollutant itself and as a precursor to secondary pollutants like ozone and $PM_{2.5}$, NO_x emissions have been subject to national emission control policies around the world."

p.4, model description: What inventory is used for aircraft emissions?

We have added the following details of the aircraft emissions inventory to Section 2.1:

> "Aircraft emissions of NO_x are from the Aviation Emissions Inventory Code (AEIC) 2019 inventory, based on Simone et al. (2013)."

p.4, line 117: Replace GEOS-Chem URL with the citation for the relevant Zenodo site used in the "Data Availability" statement.

We have added a reference to the Zenodo site used.

p.5, lines 140-141: Provide a reason for not using the modified Luo et al. wet deposition scheme.

We elected to use the standard wet deposition scheme as the modified scheme is an active area of research and still undergoing refinement (Luo et al., 2019, 2020; Luo and Yu, 2023). We also note in Section 3.1 that the alternative deposition scheme does not lead to an unambiguous improvement in performance, as demonstrated over the continental United States by Dutta and Heald (2023). We do not believe it is necessary to provide further justification in the text for using the model default scheme.

p.6, lines 174-175: This sentence is unclear as written. How are the "small number of points" determined? What is the "recommended range"? Which "two parameterizations" are these, as only one (PSS) is given?

We have rewritten this sentence as follows:

> "Burkholder et al. (2019) note that R_1 exhibits some non-Arrhenius behavior, described between 204 and 440 K. However, for simplicity, we elect to use the Arrhenius parameterization for k_1 (recommended for $195\ K < T < 443\ K$), as a small number of points (<1%) above 9 km are sampled at temperatures below 204 K. The two parameterizations differ by on average 5% (at most 15%) above 204 K."

p.6, line 185: How does the "filter out plumes" screening differ from the "fresh NOx emissions" filter?

We have edited the text to clarify that the "fresh NO_x emissions" filter uses the $NO:NO_y$

ratio, and the plume filter is a more general top 3 percentile filter used for all species.

p.7, Table 1: Convention is for table title to appear above the table, not below.
Fixed. Also fixed for Table 2.

p.8, line 210: “shows a comparison” can simply be “compares”. Similarly, “shows a summary of” can simply be “summarizes” (p. 10, line 252).
Fixed.

p.9, Figure 2: Add timings of the individual campaigns for ease of following along. These are in Figure S1, but would be helpful to include in Figure 2 too.
Added timings to subplot titles in Figure 2.

p.9, line 247: Presumably aircraft and ships are mentioned because of the functionality of the diagnostics package in the model. Would be worth mentioning this, as otherwise it seems odd that these particular anthropogenic sources are mentioned and not others that make greater contributions (road vehicles, energy generation).
We have modified the text to clarify this detail:

> “Anthropogenic sources (including here aircraft and ship emissions reported by the model’s emissions diagnostics) contribute 63% of NO_x emissions (33.9 Tg N yr^{-1})”

p.11, line 275: delete “of ” in “as well as their ratio of at the”
Fixed.

p.11, lines 280-281: What led to the conclusion that this lightning NOx and not dry season biomass burning NOx? Is this from investigating the modelled ratios of NOx:NOy in JJA and DJF, showing enhancements in the hemisphere where lightning occurs (rainy season) as opposed to where biomass burning occurs (dry season)?
The reviewer is correct that this is largely based on the spatial distributions of lightning NO_x emissions and biomass burning emissions in the Southern Hemisphere, where we see that regions of higher lightning emissions (peaking Oct-Feb) appeared to correlate well with elevated NO_x:NO_y aloft (peak in Oct-March), as opposed to biomass burning emissions (peaking July-Sept). We do not preclude a possible contribution from biomass burning, but specify that the enhancements are “primarily due to lightning”.

p.14, line 320: Given the large production and loss MPNs have a large production and loss, is it worth acknowledging the need to measure this? And under what conditions? And in what locations, based on your budget?

The formation/loss rate of MPN is so high because it is less stable than PAN at most temperatures. However, it only results in net MPN formation in the UTLS, where concentrations peak in our simulation. While more work to explore this (and many other NO_y species!) could improve the overall representation of NO_y cycling in the troposphere, we do not think this is a leading source of uncertainty in the NO_y budget, so do not make an explicit recommendation for measurements of this species.

p.16, Figure 7: Inclusion of addition NOy compounds for GEOS-Chem only suggests these aren't simulated by the other models. Is this the case? Not stated in the manuscript. If the case, say so in manuscript. If not the case, consider consider removing from Figure 7 or providing context for the reason for including these in Figure 7.

Thank you for pointing this out, we did not intend for our caption to be misleading! We have now clarified in the caption:

> "We note that the ACCMIP models represent NO_y chemistry with varying complexity but all models simulated additional NO_y species beyond the four major species shown here; however, these additional species are not represented here as their burdens were not characterized in Murray et al."

We also add to the underline text (Line 455):

> "Figure 7 shows the decadal-average burdens of four key NO_y species (NO, NO_2, HNO_3, and PAN) across ACCMIP models for the 2000s from Murray et al., (2021), compared to our simulation of 2016 where we comprehensively characterize all the forms of NO_y in GEOS-Chem (GC14)."

p.17, line 401: Typo in "issues in the simulation HNO3 and"

Fixed.

References:

Bradshaw et al. (1998) doi:10.1029/98JD00621.
Emmons et al. (1997) doi:10.1016/s1352-2310(96)00334-2.
Horner et al. (2024) doi:10.5194/acp-24-13047-2024.
Jaegle et al. (1998) doi:10.1029/97gl03591.
Lee et al. (2025) doi:10.1029/2025GL115001.
Wei et al. (2025) doi:10.5194/acp-25-7925-2025.

Reviewer 2

General Description and Recommendation:

Dutta et al. present a comprehensive overview of the current understanding of the global budget of NOy as represented in the GEOS-Chem atmospheric chemistry model. The authors first compare the GEOS-Chem NOy simulation to observations from the ATom aircraft campaign, and then examine the global burden of key NOy species, quantify the reaction pathways controlling the cycling among the major NOy species, and identify the chemical processes that need improved representation in the model. In particular, the authors highlight the importance of organic nitrogen chemistry and the photolysis of particulate nitrate, and the need to better characterize these processes.

This is a valuable and timely study. NOy plays a central role in tropospheric chemistry and has been represented in global models since the 1970's, although substantial differences among models persist. The representation of NOy chemistry in models has also evolved significantly in the past decade, with increasingly detailed treatment of organic nitrogen chemistry, heterogenous chemistry, and tropospheric halogen chemistry. The manuscript is well-written, the discussion is insightful, and the results are presented clearly through high-quality figures.

Specific Comments:

The manuscript will benefit from a more critical assessment of the representation of NOy chemistry and deposition in GEOS-Chem in the context of the the discussion of the NOy burden and chemical cycling (Sections 3.2 and 3.3). While some critical discussion of the model's representation of organic nitrogen chemistry is provided, this could be expanded to address other species, such as:

- HNO3: GEOS-Chem and many other models persistently overestimate HNO3 and it would be useful to discuss the implications of this bias for the global NOy budget.

The coupling of the NO_y system makes it challenging to identify how biases in individual species can be ameliorated. In the particular case of HNO_3, there is no obvious chemical formation or loss pathway for HNO_3 which could be altered that would not exacerbate the bias in other species. Indeed, this is why increasing wet deposition has been proposed as a possible fix. However, as we pointed out in previous work (Dutta and Heald, 2023) this leads to overestimation of observed nitrogen deposition. Another possible such pathway would be larger uptake of HNO_3 on coarse-mode aerosol. However, we have found that uptake of HNO_3 on dust for example, only produces local reductions in HNO_3. We have added the following text to Section 4:

> "In Section 3.1, we noted the development of alternative wet deposition schemes that seek to address the model's high-biased representation of HNO_3. However, these changes are not an unambiguous improvement over the existing scheme, with regional biases in concentration and deposition fluxes persisting (Dutta and Heald, 2023). Thus, investigation of additional loss processes is needed to improve the HNO_3 simulation. These may include heterogeneous processes such as the uptake of nitric acid on coarse mode aerosol (e.g. dust), though current optional representations of this process in GEOS-Chem produce only local modest reductions in HNO_3."

- Halogen nitrates: The largest flux between NOx and NOz passes through halogen nitrates, yet there is very little discussion of the relative importance of the halogen radicals involved or of the uncertainties in halogen chemistry.

Although it is true that short-lived halogens do account for large fluxes between NO_y species, on a global scale they appear to largely function as intermediates between NO_2 and NO_3 radicals. Only 5.6 Tg N yr^{-1} results in the formation of HNO_3. However, their high reactivity likely means that they are important in determining the abundance of oxidants and the processing of NO_x locally. This chemistry is still uncertain and requires measurement of a larger number of species at a greater spatial density to properly constrain. We have added the following text to Section 4:

> "A recent review by Saiz-Lopez et al. (2025) notes the need to increase the spatial and temporal density of observations of short-lived halogens, due to their high reactivity, as well as the need to include measurements of a greater variety of short-lived halogen species. The current implementation of halogen chemistry varies widely across models, largely due to limitations in our current understanding of the underlying processes."

Technical comments:

- Line 128: Coarse-mode nitrate in GEOS-Chem is also formed by the titration of alkalinity in fresh sea salt aerosols by HNO3.

We have edited the text as follows:

> "Coarse mode nitrate aerosol (NITs) is formed by the titration of sea salt alkalinity by nitric acid to form $NaNO_3$."

- Line 236: Please specify the months of the ATom deployments.

We have added deployment dates below the campaign labels.

- Line 250: Not all emissions in the table are from CEDS.

We have removed the reference to CEDS. Details of emissions inventories are in Section 2.1.

- Table 2: For completeness, please include the stratospheric source of HNO3 in the table.

Table 2 quantifies only direct emissions of NO_y, thus transport would not be an appropriate addition. We note the role of transport in balancing the HNO_3 budget in Section 3.2.

- Line 300: Should this read “aerosol organic nitrates”?

Yes, thank you!

- Line 365: The NO2+NO3+M → N2O5+M reaction is termolecular.

We have changed “pressure-dependent bimolecular” to “termolecular”.

- Line 368: Should this read ClNO2 instead of ClNO3?

Yes, great catch!

References

Burkholder, J. B., Sander, S. P., Abbatt, J., Barker, J. R., Cappa, C., Crounse, J. D., Dibble, T. S., Huie, R. E., Kolb, C. E., Kurylo, M. J., Orkin, V. L., Percival, C. J., Wilmouth, D. M., and Wine, P. H.: Chemical Kinetics and Photochemical Data for Use in Atmospheric Studies, Evaluation No. 19, Jet Propulsion Laboratory, Pasadena, CA, 2019.

Dutta, I. and Heald, C. L.: Exploring Deposition Observations of Oxidized Sulfur and Nitrogen as a Constraint on Emissions in the United States, Journal of Geophysical Research: Atmospheres, 128, e2023JD039610, https://doi.org/10.1029/2023JD039610, 2023.

Fisher, J. A., Jacob, D. J., Travis, K. R., Kim, P. S., Marais, E. A., Chan Miller, C., Yu, K., Zhu, L., Yantosca, R. M., Sulprizio, M. P., Mao, J., Wennberg, P. O., Crounse, J. D., Teng, A. P., Nguyen, T. B., St. Clair, J. M., Cohen, R. C., Romer, P., Nault, B. A., Wooldridge, P. J., Jimenez, J. L., Campuzano-Jost, P., Day, D. A., Hu, W., Shepson, P. B., Xiong, F., Blake, D. R., Goldstein, A. H., Misztal, P. K., Hanisco, T. F., Wolfe, G. M., Ryerson, T. B., Wisthaler, A., and Mikoviny, T.: Organic nitrate chemistry and its implications for nitrogen budgets in an isoprene- and monoterpene-rich atmosphere: constraints from aircraft (SEAC^{4}RS) and ground-based (SOAS) observations in the Southeast US, Atmospheric Chemistry and Physics, 16, 5969–5991, https://doi.org/10.5194/acp-16-5969-2016, 2016.

Horner, R. P., Marais, E. A., Wei, N., Ryan, R. G., and Shah, V.: Vertical profiles of global tropospheric nitrogen dioxide (NO_2) obtained by cloud slicing the TROPOspheric Monitoring Instrument (TROPOMI), Atmospheric Chemistry and Physics, 24, 13047–13064, https://doi.org/10.5194/acp-24-13047-2024, 2024.

Luo, G. and Yu, F.: Impact of Air Refreshing and Cloud Ice Uptake Limitations on Vertical Profiles and Wet Depositions of Nitrate, Ammonium, and Sulfate, Geophysical Research Letters, 50, e2023GL104258, https://doi.org/10.1029/2023GL104258, 2023.

Luo, G., Yu, F., and Schwab, J.: Revised treatment of wet scavenging processes dramatically improves GEOS-Chem 12.0.0 simulations of surface nitric acid, nitrate, and ammonium over the United States, Geoscientific Model Development, 12, 3439–3447, https://doi.org/10.5194/gmd-12-3439-2019, 2019.

Luo, G., Yu, F., and Moch, J. M.: Further improvement of wet process treatments in GEOS-Chem v12.6.0: impact on global distributions of aerosols and aerosol precursors, Geoscientific Model Development, 13, 2879–2903, https://doi.org/10.5194/gmd-13-2879-2020, 2020.

Murray, L. T., Fiore, A. M., Shindell, D. T., Naik, V., and Horowitz, L. W.: Large uncertainties in global hydroxyl projections tied to fate of reactive nitrogen and carbon, Proceedings of the National Academy of Sciences, 118, e2115204118, https://doi.org/10.1073/pnas.2115204118, 2021.

Saiz-Lopez, A., Mahajan, A. S., Abbatt, J., Bobrowski, N., Brown, S. S., Burrows, J. P., Carpenter, L. J., Chipperfield, M. P., Cuevas, C. A., Fernandez, R. P., Hossaini, R., Kinnison, D. E., Lamarque, J.-F., Finlayson-Pitts, B. J., Plane, J. M. C., Platt, U., Pratt, K. A., Ravishankara, A. R., Salawitch, R. J., Saltzman, E. S., Simpson, W. R., Solomon, S., Thornton, J. A., and Wang, T.: The influence of short-lived halogens on atmospheric chemistry and climate, Nature, 648, 289–299, https://doi.org/10.1038/s41586-025-09753-x, 2025.

Wei, N., Marais, E. A., Lu, G., Ryan, R. G., and Sauvage, B.: Characterization of reactive oxidized nitrogen in the global upper troposphere using recent and historic commercial and research aircraft campaigns and GEOS-Chem, Atmospheric Chemistry and Physics, 25, 7925–7940, https://doi.org/10.5194/acp-25-7925-2025, 2025.