Review of manuscript titled "Exploring the sensitivity of atmospheric nitrate concentrations to nitric acid uptake rate using the Met Office's Unified Model" by Jones et al. submitted to Atmospheric Chemistry and Physics Discussions

This work presents the UK Met office's Unified Model (UM) with a new nitrate scheme in which sensitivity of thermodynamic equilibrium for NH_4NO_3 production to the HNO_3 and NH_3 uptake to aerosol, is explored. Furthermore, the impact of these sensitivities on mass budget and total deposition rates for NH_4, NO_3 (fine and coarse), NH_3 and HNO_3, $PM_{2.5}$, AOD and radiation changes etc. have been descriptively presented on a global scale. This work specifically provides a way to address overestimation of the NO_3 aerosol by current crop of climate models, in a computationally efficient way.

In general, the manuscript is quite descriptive in terms of presenting the methods with governing equations (including supplementary material), which is valuable for reproducibility, and provides descriptive robust analysis and validation of its findings with observation networks across the globe (temporal and spatial comparisons), and previously reported aerosol budgets and lifetime. The work presented in this manuscript is no doubt quite robust and essential to pave way for more complex parametrizations to be added in future to UM. The introduction of this manuscript specifically can be referred to students of atmospheric chemistry as a broad overview of secondary inorganic aerosol formation and the state of their modeling (specifically NO_3 aerosols).

General Comment:

The main overarching issues were pertaining to some detailed explanations in methods and their explanatory text in main manuscript that can either be summarized, tabulated and/or shifted to supplement to allow reader(s) to get at the results and discussions quickly, and not be overwhelmed before they get to that part. The lack of brevity specifically in 'Methods' and to some extent in 'Results' needs to be addressed by the authors before a revised version of manuscript can be accepted finally for publication (some suggestions in Specific Comments). Also, the distinction between fine vs coarse NO_3 in terms of nomenclature say coarse mode nitrate as hetNO_3 can be confusing to readers (See Specific Comments on Table 1). However, I encourage this manuscript to be accepted for publication in Atmospheric Chemistry and Physics journal with minor revisions.

Specific Comments:

Lines 40-60: Make sure the paragraphs are Justified instead of left aligned and ensure the same in rest of manuscript consistently.

Lines 122-123: Edit the following as:

"The hybrid-dynamical nitrate scheme developed by Benduhn et al. (2016) in the standalone GLOMAP-mode model is currently not implemented ~~yet to successfully transition to~~ in the UM."

Lines 125-126: Edit the following as: (if this is accurate)

"…~~in order~~ to ~~fill the NH4 and NO3 shaped void.~~ address the gaps in modeled NH_4 and NO_3 with their respective observations."

Section 2.1: Will suggest the authors to consider summarizing the configuration of the UM used to test the new nitrate scheme as described in section 2.1 in tabular form either in main manuscript or even move to supplement. This will help readers to get to the focus of reader(s) quickly to the new nitrate thermodynamic scheme in UM quickly.

Table 1:

- **a)** please denote that $\boldsymbol{\sigma g}$ is referred to geometric standard deviation in the Table 1 caption.
- **b)** Can the authors enunciate more on the difference between NO_3 and hetNO_3 in accumulation and coarse mode? (Lines 209-210 ["*NH4 and NO3 mass is emitted into the Aitken and accumulation soluble modes and may be transferred to the coarse soluble mode via aerosol processing, while hetNO3 is limited to the accumulation and coarse soluble modes*."] seems to explain this? But might help to etch it out more in terms of this question and Table 1).
- **c)** In Lines **409-410:** authors state "'fine NO3' refers to NO3 associated with NH4 while 'coarse NO3' refers to NO3 associated with dust and sea-salt (i.e. NO3 in hetNO3)." Similar clarification to address comment **b)** early on in the text would be helpful.

Section 2.2.1: Please ensure chronological ordering of supplemental figures (for instance S2 comes after S3 in text now), re-arrange in the order they are mentioned in text. Make sure Tables, figures are arranged in a chronological order in general throughout the manuscript.

Lines 279-281: *"However, atmospheric Aitken mode number concentrations generally exceed accumulation mode concentrations, particularly over populous land regions and increasingly with altitude."*
Referring to the above statement, did the authors observe any converse trend (i.e. higher accumulation ode concentrations) over say rural or coastal non-urban regions? Discussing the spatial patterns in Fig S4 briefly in 1-2 sentence(s) would be a valuable inference to add from the manuscript's findings.

Lines 294-295: Apart from Wang et al., 2020 (Nature) showing NH_4NO_3 nucleating for new particle formation (NPF), there has been an increasing interest in exploring NPF parametrizations specifically for bridging the model-observation number concentration gaps at high elevations (https://agupubs.onlinelibrary.wiley.com/doi/10.1029/2018JD029356) . It would be good to point this out may be in discussions on future steps (in conclusions), as NPF parametrizations are still an evolving area when it comes to global climate models.

Lines 307-309 (see next comment as well on Section 4): Further/clearer explanations on the merit of following assumptions would be better:

"Additionally, dust is assumed to uniformly constitute 5 % Ca^{2+} by mass, which differs from the approach in Remy et al. (2019) who used a spatially hetereogeneous Ca^{2+} fraction. Dust alkalinity is titrated by uptake of HNO_3 until the dust pH is neutralised whereupon HNO_3 stops condensing, while no such limitation is applied for sea-salt."

Lines 390-400 (and Section 4): Follow up in conclusion on how uncertainty in different emission sources specifically NH_3 with overestimated inventory can be a major source of uncertainty.
I see points in Section 4 has been made regarding the NH_3 and NO_x inventory-induced uncertainty and N_2O_5 chemistry simplification (Lines 735:738: "*An accurate NH3 and NOx emissions inventory is vital for a proficient simulation of NH_4 and NO_3 concentrations. HNO_3 concentrations also appear to be overestimated over the western US (Fig. 5) in these simulations, which may emanate from an oversimplification of heterogeneous N_2O_5 chemistry in UKCA Strattrop1.0, given that the uptake coefficient in that reaction is uniformly set to 0.1 (Archibald et al., 2020).*"]. But similar impacts from say assumptions made in Lines 307-309 (for instance) can be further dissected in conclusions as they are missing.

Figure 3: The only fundamental conceptual critique I have on result section is pertaining to Fig. 3: is that the manuscript does not elucidate much on the 'role of Convection' in their version of UM model that can also bring nucleation precursors (NH_4, NO_3 for instance) from the ground level to the free troposphere? The vertical profile obviously is limited to 0-6 km as NPF or nucleation has not been included (see comment for **Lines 294-295**). This limitation is essential to be mentioned as stated earlier, albeit as a future step if beyond the scope of current manuscript.

Figs 5,6 and 8: The discussions around these figures seem to give similar inferences and can be synthesized together in terms of their summary pointing to similar inferences on FAST introducing positive biases etc. Its understandable than Figs. 5, 6 are annual means and Fig. 8 gives a seasonal variation, but still can be very much summarized together. Or some parts of them can be moved to supplement.

Figure 11: Authors can do away with Panels (a),(b), and (e) and their discussions on AOD results for CNTL, FAST and SLOW sensitivity runs, and just show difference maps (c, f). Can move the Panels (a),(b),(d),(e) as separate figure into supplement and summarize the model vs observed AOD in result text, referring to the new supplemental figure. There is a need to synthesize the result section, in terms of what take-out messages the authors want to stand out to the reader(s).