Overall Summary:

This work describes a new thermodynamic equilibrium scheme developed, which explicitly considers kinetic limitations of HNO_3 and NH_3 uptake to aerosols in the UK's climate model. They test the common assumption in global models that NH_4NO_3 concentrations reach thermodynamic equilibrium instantaneously, by varying the assumed the heterogenous uptake rate of HNO_3 and NH_3 in the model and investigating the impacts on important climate parameters such as $PM_{2.5}$ concentrations, AOD, radiative balance, etc. They demonstrate that such a variation does have significant air quality and climate implications, and that the computational cost of implementing such a scheme is worthwhile. But, perhaps most significantly, they highlight the high sensitivity of ammonium nitrate concentrations to nitric acid uptake rates and, therefore, provide a mechanism for reducing nitrate concentrations biases in other climate models, which make similar instantaneous thermodynamic equilibrium assumptions.

Given the robustness of this problem in climate models beyond just this specific model, this paper represents a substantial contribution to scientific progress well within the scope of Atmospheric Chemistry and Physics. In this paper, the methods are clearly outlined, the conclusions reached are well supported, the abstract is clear, and importantly, even though the source code for the model cannot be provided due to licensing, the details in the paper and supplement are enough that the methods outlined could be reproduced in other models. The citations are appropriate and worth commending the authors for their efforts as it is obvious, they have a detailed understanding of the body of literature on this topic. The title is appropriately descriptive, and overall, the paper is very readable. I did find the number of figures to be overwhelming and excessive in some places and have suggested a few ways to make the results section slightly clearer/ potentially shorter, which I believe would increase the reach and impact of the paper. However, overall, I recommend that the paper be accepted with minor revisions.

General Comments:

The abstract, introduction, background, motivation, methodology, and conclusions of the paper are particularly well written - perhaps one of the best of papers I have reviewed. However, I found the results section of the paper overwhelming, at times, given the number of figures, the number of panels in figures, and the amount of information contained in any one figure. In stark contrast to the readability of the rest of the paper, I felt that the results section could be significantly improved by (1) the addition of a single summary sentence at the end of many paragraphs in order to better highlight conclusion / context of the conclusion that is reached and (2) by tightening up the figures (e.g. reducing the total number of figures, reducing the number of panels, or reducing the number of lines on any one figure). To be clear, I found the conclusions and abstract to be very understandable and well supported. However, I thought the communication of the points within the results section itself- particularly how the text discussion of figures is structured- could be improved. In the specific comments I've tried to point out both where this is done well and where, even as a "target reader" I initially struggled to follow.

One question I did walk away from the paper with, is whether the testing of the model's sensitivity to instantaneous equilibrium was truly novel? I am curious as to whether this is the first paper exploring this or whether it is something that was implemented in the UM following the implementation of something similar in a different model. I'm not familiar enough with what's done in models beyond GEOS-Chem (which uses ISORROPIA II & therefore the instantaneous equilibrium assumption), but it strikes me that how novel this approach is this should be made more clear in the final paper- either in order to point out the truly novel result of this work, or to acknowledge that which came before in a way that is more explicit than is done now.

As a small final note, I found it generally confusing that the authors refer to coarse mode nitrate as hetNO3. The terms are used interchangeably in the main text and discussion, but I think just changing the moniker hetNO3 to something like coarse$NO_{3,}$ c$NO_{3,}$ bigNO_3 would be less confusing. Simply CNTRL+F'ing the text to ensure consistency in their reference to this would certainly improve the communication of the result.

Specific Comments:

Line 123: …"model is yet to successfully"- Typo: I think "yet" should be "set"

Line 176-177: I highlighted this line because on my initial read through, I was concerned that the assumption of a static cloud pH could impact the results contained within. I saw much later in the discussion that this assumption was addressed- it may be worth mentioning that you address this later in Section XX.

Lines 230-235 and 285-296: I found myself struggling here to translate some of the mathematical statements to readable words. Instead of stating "$Ta*TN > K_p$" or "$Ta*TN < K_p$" , I suggest the authors put what this means into physical context using words for the readers. For example: "If available nitrate & ammonium suggest the equilibrium of Reaction 2 is in the forwards direction, thereby promoting the condensation of HNO_3 and NH_3 to form NH_4NO_3 ($Ta*TN > K_p$), then the equilibrium concentration of NH_4NO_3 it is solved using Eq 3. Otherwise…"

Lines 293-295: I was confused about the assumption that ammonium nitrate does not nucleate new particles and thought some elaboration was needed here. There's a recent Nature paper showing that ammonium nitrate out of equilibrium in clouds can be important nucleation/growth events even if such a state is only reached for a few minutes (https://www.nature.com/articles/s41586-020-2270-4). This is likely less important within a global climate model, but it is worth at least a sentence or two of elaboration.

Line 309: Why is alkalinity not titrated for sea-salt? Is it just an assumption that HNO_3 is in excess of Na^+? I found it odd that this was stated with no reasoning.

Line 389: It seems to me that the uncertainty in emissions driving the model are a major contributor to the discrepancies between the model and observation comparisons made in the results section- which is why there is time spent making seasonal and spatial comparisons between the model and observation networks. I think its worth a sentence here preparing the reader for that pointing out the significant of these emissions uncertainties- something along the lines of: "Such large discrepancies in NH_3 emissions inventories can impact direct model- measurement comparisons … which make it important to consider the spatial and temporal trends rather than just the overall magnitudes. For this work exploring the sensitivity of NH_4NO_3 to thermodynamic equilibrium assumptions, the direct comparison of model performance to observations is done with the goal of understanding the degree to which thermodynamic assumptions may push the model out of realistic behavior rather than best re-creating observations"…

Line 419-420: I wanted to highlight these lines, because I they do a really good job of "summarizing" what the previous paragraph states / contextualizing the results pointed out. It is what I was longing for in other parts of the results section. I suggest the authors look through the other final sentences in the results section and model them after this paragraph.

Lines 421-434: This is one of the results paragraphs that is not well contextualized to the physical chemistry occurring/ the overall point of the results communicated. The authors point out the differences in the fast and slow fine and coarse model NO_3 sources and burdens, coarse fractions & how they compare to other models. But they leap to " these results… corroborates the assertion that NH4 and NO_3 aerosol production is at the upper end of efficiency & that NH_3 is limiting" without clearly explaining why. (e.g. Fine mode production of NO_3 is faster in FAST (ok, duh), but coarse mode production of NO3 is faster in SLOW (why physically? Is it that there is more HNO_3 in the gas phase because NH_4NO_3 production is slower, so that more HNO_3 is available to condense on the coarse mode particles?) Then contextualize WHY that the % of the NO_3 burden in the coarse mode of SLOW is closer in HA14 and FAST is closer to XP12 – what assumptions do those studies make about the HNO_3 uptake (instantaneous?). Is the % of NO_3 burden in coarse mode comparisons just to say that it's within the range of other models? That's what the final sentence seems to suggest. This just needs to be more clear- there's a sentence missing connecting the stated results to the conclusion made.

460: Nicely communicated & contextualized result.

Figure 4: This figure was particularly hard for me to decipher, and I spent far too long trying to understand the message it was communicating. The conclusion in the text is the only reason I was able to follow what this was showing. First, the key has cut off the symbol for "hetNO_3 Fast" so that its symbol looks identical to NO_3 fast- I assume that the solid line is NO_3 fast and the long dashes are hetNO_3, but the key does not distinguish them. I think this plot could be greatly improved if it was more clear which lines we're supposed to be comparing on each figure. Right now, that is done with the different line styles (e.g. compare black solid like to black solid line), but I think the line thickness are different between the fast and slow simulations, which makes identifying which lines are supposed to be comparable difficult (e.g. the dashed black line looks totally different than the color dashed line). Another way to improve this figure would be to improve the key- Put the slow and fast lines that are comparable next to one another (so 2 columns in the key).

Line 499-506: Clarity of result could be improved/ needs a better summary sentence.

Line 518: The authors mention several times that the "model output is linearly interpolated to measurement sites", but its not totally clear exactly what that means. Is it that value assumed at the specific lat/long of the site is actually interpolated from its distance to other grid boxes & their values?

Line 525-533: This is another section where I was looking for a better summary sentence. I'm not sure if the point of these figures is to show that FAST/SLOW is "better" at simulating observations or that simply changing the assumption about thermodynamic equilibrium has a huge impact on NO_3, NH_4, and HNO_3. Figure 5 and 6 are pushing me to decide which assumption is "better"- but the emissions bias seems to muddle making a direct conclusion. E.g. SLOW looks closer to the 1:1 line, but it's only there because of emissions biases? Does this mean FAST is better? Is that the point of the figure? Perhaps you could add a "theoretical" 1:1 line to show what you'd expect if there weren't emissions biases? The conclusions section describing this result is much clearer (lines 710-715) than the results discussion here.

Line 597-599: I was also wondering about N_2O_5 chemistry being promoted in the winter & how that is simulated in the model? Perhaps worth a mention here as a reason for the PM2.5 disagreements. (Maybe see Shah et al., 2018: https://www.pnas.org/content/115/32/8110/tab-article-info).

Figure 10 & Section 3.4: I'm not sure that Figure 10 adds anything to the paper that cannot be described in the text. The figure has so many panels that it's very confusing to read and hard to compare the panels needed to get to the in-text conclusions. I suggest the authors consider moving it to the supplement or simplifying the figure.

Figure 12: Given the small region where the TOA radiation differences are significant between the two results, I also suggest cutting this figure or moving it to the supplement. The results can be adequately described in the text without a figure.