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Interactive comment on “The contribution of plume-scale nucleation to global and regional aerosol and CCN concentrations: evaluation and sensitivity to emissions changes” by R. G. Stevens and J. R. Pierce

Anonymous Referee #1

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1 Review

The paper describes the implementation of a new sub-grid sulphate parameterization (P6) into the global chemical-transport and aerosol microphysics model GEOS-Chem-TOMAS. The parameterization is compared with two other parameterizations of sub-grid scale sulphate, in a variety of model scenarios. Although the paper cannot definitively state that the P6 parameterization provides better agreement with aerosol

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observations, the authors believe that it can better account for spatial heterogeneity than the other parameterizations tested. The parameterization has potential, and future iterations may account for other processes highlighted in the paper, such as the effects of anthropogenically controlled SOA on sub-grid new-particle formation and growth. The findings of the paper are within the scope of ACP, and it should be published after minor revisions.

2 Consistent grammatical issues

Two grammatical issues should be consistently addressed within the paper for ease of reading. The first is the frequent nominalisation of increase and decrease. While the use of the noun form instead of the verb form is not incorrect, sentences can often be structured more clearly by using the verb form. The second is the repeated use of the word “thus”. I would recommend the use of “therefore” in the majority of cases. While the use of “thus” as a synonym of “therefore” is often accepted in general language, it is technically an adverb and should primarily be used in scientific writing when describing how an action was performed, rather than to mean “as a result of...”.

Abstract

P 21474

L 8: move “in simulations using P6 sub-grid sulphate” to before “of particle number concentration”.

L 10-13: Nominalisation of “increases” makes this sentence more complicated. I recommend rewording: “For global increases of 50 % in emissions

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of either SO_2 or NO_x , or both SO_2 and NO_x , we find that globally, annually averaged N80 increase by 9.00%, 1.47%, or 10.24% respectively. However, both sub-grid and grid-resolved processes contribute to these changes."

P 21475

L 17: "Momentarily" is an ambiguous word. I would recommend removing it.

P 21476

L 6-9: This sentence should be split into two shorter sentences. The phrase "hundreds and tends of kilometres or more, respectively" is confusing and should be reworded.

P 21477

L 1, 3: There is some ambiguity in saying "increases by X% to Y%" - I am pretty sure you mean "increases by between X and Y%", in which case I would recommend using that phrasing.

L 5: Add a colon after "sulphate".

L 5-8: Is all remaining sulphur mass condensed onto pre-existing particles, or is a certain proportion of sulphur calculated as being condensed, and 5% of this is emitted directly into the nucleation mode?

L 10-11: Change the verb increase from passive to active voice - "CCN increased by up to..."

L 19: Due to the frequent (and necessary) use of the word "uncertainty" in this paragraph, I would recommend "uncertainties in CCN concentration as those of SO_2 emissions".

L 23-24: The number of the uncertainty range is not relevant to the paper and should be removed for clarity. I would recommend cutting the sentence after "tested".

L 26: Move the phrase "for the diameter of sub-grid-sulphate particles" to between "values" and "used".

L 27-: Rephrase. I would suggest cutting "due to uncertainties in sub-grid sulphate" and replace "compared to the range of estimates" with "as".

P21479

L 8: Replace "due to" with "in the form of/via" or other appropriate term.

L 10-12: Rephrase sentence.

L 17: I'm honestly not sure that adjoint is the appropriate word, but I also can't think of a suitable one to suggest.

P 21480

L 10: A verb has been omitted - "it overwritten".

P 21481

L 9: An article has been omitted - "inherits limitations of".

L 11-12: For clarity, relocate "due to...(HONO)" to between "formation that".

L 12-18: Is the aqueous-phase pathway still accounted for in GEOS-Chem-TOMAS in general, even if it is omitted from the P6 parameterization?

L22-23: This sentence would be clearer if phrased in the passive voice (admittedly a rare occurrence!).

P 21482

L 28: Add “and described below” to the end of the sentence - the Table does not stand alone if the reader moves there directly. There will be more discussion of tables and figures later in the review.

P 21484

L 18-20: While technically correct, the word “greater” followed by “decreases” in the next sentence is counterintuitive. Similarly, in lines 27-28, “increased globally” followed by “decreased globally” disrupts the flow of reading.

P 21485

L15-18: Split into 2-3 short sentences.

P 21487

L 22: Fewer, not less.

P 21488

Do you intend to account for the preferential formation of SOA within anthropogenic plumes in the future? Do you expect the effect to be significant?

L 19: Dampen, not dampens.

L 24: Split the phrase “(as) shown in Table 2” into a separate phrase with a comma.

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P 21489

Could you estimate the difference in radiative effect between the three parameterizations?

P 21490

Would it be possible to support the assertion that non-linearities in the P6-adjoint equations will not quantitatively affect the findings of the analysis? Perhaps via a set of Monte Carlo simulations or an explicit comparison between the average of a month's simulations versus the simulation using a month of averages?

L 18: Move the mention of Fig. 5 to after the discussion of Fig. 4 - preferably P 21491, after end of first paragraph.

L 25-27: The second half of the sentence feels like a non sequitor and may not be necessary.

P 21491

L 4-9: This is a long, confusing sentence. At the least, I would recommend putting brackets around (since the remaining sulphate in P6 is emitted into the Aitken mode and not condensed onto pre-existing particles).

L16: Put the Fig. 5 description at the end of this paragraph.

L 16-19: The CS panel in Fig. 5 is uniformly blue. Is this because it is constant, or just because it doesn't change outside of the 1-2 % $\%^{-1}$ range?

P 21492

L 19-20: specify annually averaged differences in N_{80} are shown in Fig. 7.

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P 21493

L 3-9: These two sentences need to be revised. I would suggest, "Increases in the background concentrations of SO_2 (bgSO_2) and NO_x (bgNO_x) in the P6_hiSO2 and P6_hiNOx simulations, respectively, will lead to differences in the P6 outputs. The resulting changes in sulphate formation and growth (at both the grid-resolved and sub-grid scales) will result in changes to the grid-resolved aerosol condensation sink(CS), which will also influence the P6 outputs."

L 22: "number of" or "proportion of" sub-grid sulphate particles?

P 21497

L 26: than, not that.

This section really suffers from the nominalisation of increase and decrease. I have suggested some revisions below; feel free to use other ones.

P 21498

L 7-10: Including an additional 100 Tg yr^{-1} of SOA led to an increase in N80, attributable to condensational growth of sub-grid sulphate particles.

L 15-18: The additional SOA increased the pre-existing condensation sink, drastically decreasing the sub-grid new-particle formation predicted by the P6 parameterization, and decreasing the inclusion of sub-grid sulphate on N80.

L 24-L7 of P 21499: We found that for a global increase in emissions of 50%, globally, annually averaged N80 increased by 9.00% (SO_2), 1.47% (NO_x), or 10.24% (SO_2 and NO_x). Both the size and number of sub-grid sulphate particles emitted increased with increasing SO_2 emissions, as did the concentration of SO_2 available to form SO_4 . Both sub-grid and grid-resolved oxidation of SO_2 increase

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with increasing NO_x emissions, except in very polluted regions ($[\text{NO}_x] > \sim 1$ ppb), where oxidation rates increase with decreasing NO_x concentrations. Both the number and size of sub-grid particles decrease with increasing NO_x emissions. When both O_2 and NO_x emissions increase, the number of sub-grid sulphate particles emission decreases over polluted regions and increases over remote regions, and there is little change in the size of sub-grid sulphate particles.

P 21499

L 11-13: For cases without anthropogenically controlled SOA, the absolute log-mean bias between simulated and observed number concentrations was reduced by including any sub-grid sulphate scheme.

L 15-17: The P6 sub-grid sulphate scheme only slightly altered/changed the absolute log-mean bias from the case with no sub-grid sulphate.

L 22-23: Redundancy of also/as well.

P 21500

L 3: "Thus" to "therefore".

Figures:

Table 1: Break the table up into sets of four (plus the three at the end) using horizontal lines. Add references for AS3, LY5, P6 to the caption so that the Table and its caption can be understood independently of the text.

Table 2: Re-group the simulations into four groups of three instead of three groups of four - I would also recommend reordering as yXSOA_Napa, nX-SOA_Napa, yXSOA_Act, nXSOA_Act. It will be easier to compare the difference parameterizations then. Include horizontal lines between groups if kept in the format of a table; however, consider reformatting this as a figure.

Table 3: Mention in the caption that the decreases in N3 and N10 are down to less nucleation or more coagulation, while the increases are from more nucleation; while the increases in N40 and N80 are from new sub-grid-scale particles.

Table 4: The best results have not been bolded or italicized.

Figure 1: Add the units to either the colour bar or the caption (not just the title).

Figure 4: Emitted particles, not particle.

Figure 8: Add column and row titles in larger font (SO_2 , N_{new} , etc.). In Fig. 8 (f), is the decrease in diameter due to more smaller particles or fewer larger particles?

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