We would like to thank both referees for their helpful comments, which have served to improve the paper. In order to respond to the referees' comments, we quote both reviews below. We italicise the referees' comments for clarity. For brevity, we have omitted from this response the technical corrections suggested by the referees where the recommended change to the manuscript was straightforward and implemented in our revised version, so we do not explicitly repeat them here. In such cases, we agree with the referee that the suggested change improves the paper, and we have made the changes exactly as recommended.

Referee #1:

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 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...".

In addition to the specific changes the referee has suggested, we have made changes throughout the manuscript to more clearly phrase sentences containing the words “increase” or “decrease”. We have also replaced all instances of the word “thus” with “therefore”, “hence”, or “consequently”.

Abstract

P 21476

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

We have rephrased the sentence to the following:

"Currently, the resolutions of regional- and global-scale models are typically at least tens or hundreds of kilometres, respectively. These models are therefore unable to accurately resolve the formation and growth of aerosols within these plumes using grid-box averages for chemical concentrations, aerosol concentrations, and meteorological values."

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.

We have rephrased the sentence to the following:

"Additionally, they found that CCN(0.2%) increased by either 23% or 53% averaged over the global boundary layer, and that the aerosol indirect effect radiative forcing increased by either 11% or 31% (depending on the grid-resolved nucleation scheme used in the boundary layer)."

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?

Of the emitted SO_2 that is assumed to form sulphate on the sub-grid scale, 5% is emitted directly into the nucleation mode and the remaining sub-grid sulphate is condensed onto pre-existing particles. We have rephrased the sentence to the following:

"The study of Yu and Luo (2009) used yet another approach for representing sub-grid sulphate: of the emitted SO_2 assumed to form sulphate on the sub-grid scale, 5% of sulphur mass is emitted directly into the nucleation mode described above and the remaining mass is condensed onto the existing accumulation-mode particles."

P21479

L 10-12: Rephrase sentence.

We have rephrased the sentence to the following:

"In order to better understand the effects of pollution controls on CCN concentrations, we also investigate the sensitivity of the N80 enhancement from sub-grid sulphate to greater emissions of SO_2 and NO_x"

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.

We agree with the referee that "adjoint" is not the most appropriate term. We therefore have replaced references to the P6 adjoint with P6 Gradient Subroutine throughout the text.

P 21481

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?

The aqueous-phase pathway is indeed accounted for at the grid-resolved scale by GEOS-Chem-TOMAS. We have added the following sentence for clarity:

"We note that at the grid-resolved scale, the GEOS-Chem-TOMAS model does represent aqueous

oxidation of SO_2, in-cloud aerosol processing, and condensational growth of aerosol due SOA."

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

We have rephrased the sentence to the following:
"We therefore expect that the variability in new-particle formation and growth rates within sulphur-rich plumes is well represented by the P6 parameterization."

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.

The first sentence has been rephrased to the following:

"Lonsdale et al. (2012) showed that the average emissions rate of SO_2 from US coal-fired power plants decreased by 36% from 1997 to 2010, and that the emissions rate of NO_x decreased by 52% from 1997 to 2010."

Note that numeric values have changed because we now state the relative difference with reference to the 1997 values, instead of the relative difference with reference to the 2010 values.

We rephrase "We note that actual SO_2 emissions have not decreased globally," to "We note that real-world SO_2 emissions were not greater in the past for all locations globally,"

P 21485

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

We have rephrased the sentence to the following:
"We present the relative changes in globally and annually averaged boundary-layer N3, N10, N40 and N80 due to sub-grid sulphate in Table 1. The values in the table are calculated by comparing each listed simulation with the corresponding simulation that had no sub-grid sulphate, the same amount of SOA emissions, and the same grid-resolved nucleation scheme. We exclude from Table 1 the emissions sensitivity studies, which will be discussed in Sect. 5."

P 21487

L 22: Fewer, not less.

We have rephrased the sentence to the following:
"In contrast to the AS3 and LY5 simulations, the enhancement in N80 due to sub-grid sulphate for the P6 simulations is smaller for the simulations where anthropogenically controlled SOA is included (yXSOA, panels a and c) than for the two simulations without this extra source of SOA (nXSOA, panels b and d)."

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?

We do intend to account for the preferential treatment of this SOA in the future. The global formation rate of anthropogenically controlled SOA indicated by Spracklen et al. (2011) is on the same order of magnitude as the global anthropogenic SO_2 emission rate, and far exceeds the gas-phase production rate of H_2SO_4 (~factor of 6). If this SOA preferentially condenses onto particles within anthropogenic plumes, including newly formed particles within these plumes, we would expect a significant effect on the growth and survival rate of these particles. We have added the following to the manuscript:

"We intend to include the effects of this SOA on sub-grid formation and growth in a future version of the P6 parameterization, once these processes become better understood."

P 21489

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

We do not currently have the ability to preform detailed radiative effect calculations on our own, although we are currently developing this capacity. We can very roughly estimate the indirect radiative effect of sub-grid sulphate as:

$$RF = -\frac{1}{3} F_{TOA} f_{cloud} T^2 A_{cloud} (1 - A_{cloud}) \ln\left(\frac{N80_{sgs}}{N80_{nosgs}}\right)$$

where F_{TOA} is the downward flux of shortwave radiation at the top of the atmosphere, f_{cloud} is the fraction of Earth's surface area covered by low clouds, T is the above-cloud transmittance of the atmosphere, A_{cloud} is the average cloud albedo, and $N80_{sgs}/N80_{nosgs}$ is the ratio between the globally averaged N80 in the simulation of interest and a simulation with no sub-grid sulphate (Seinfeld and Pandis, 2006). If we approximate F_{TOA} as 343 W m^{-2}, f_{cloud} as 0.3, T as 0.76, and A_{cloud} as 0.5, and we use the changes in globally, annually averaged surface N80 from the rightmost column of Table 2 to determine $N80_{sgs}/N80_{nosgs}$, we obtain the following:

simulation	% change in N80	indirect radiative effect[W m^{-2}]
AS3_yXSOA_Napa	+9.73	-0.46
LY5_yXSOA_Napa	+19.72	-0.89
P6_yXSOA_Napa	-0.86	+0.04
AS3_nXSOA_Napa	+4.94	-0.24
LY5_nXSOA_Napa	+10.78	-0.51
P6_nXSOA_Napa	+3.46	-0.17
AS3_yXSOA_Act	+8.43	-0.40
LY5_yXSOA_Act	+19.11	-0.87
P6_yXSOA_Act	+1.32	-0.06
AS3_nXSOA_Act	+4.07	-0.20
LY5_nXSOA_Act	+10.56	-0.50
P6_nXSOA_Act	+5.71	-0.28

Note that we have re-ordered the simulations in this table as recommended by the referee for Table 2. The difference in the indirect radiative effect between the three parameterizations ranges between about 0.3 W m^{-2} and 0.9 W m^{-2}, depending primarily on whether or not anthropogenically controlled SOA is included.

We would also expect greater radiative effects regionally, as well as greater differences in radiative effect regionally. For example, in the two simulations with AS3 sub-grid sulphate and Napari ternary nucleation, N80 increased by more than 20% over the Pacific Ocean east of northern South America. As this is a region near the equator with frequent cloud cover, we expect that the radiative effect may exceed 1 W m^{-2} in this region. The change in N80 in this region was much smaller for the corresponding P6 simulations, and N80 over the northern Pacific decreased in those simulations, which would indicate a warming over those regions. The spatial heterogeneity in shortwave radiation, cloud cover, and N80 changes, as well as the implicit assumptions about the relationship between N80 and cloud droplet number concentrations, lead to large uncertainties in the global indirect radiative effect calculated here.

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?

The monthly emissions of SO_2 (E_{SO2}) and the monthly sub-grid scale sulphate emissions (E_{SO4}) were available as output of the GEOS-Chem-TOMAS simulations. It is therefore possible to calculate the monthly mean of the fraction of SO_2 oxidized (f_{ox}) as:

$$f_{ox} = \frac{\frac{E_{SO4}}{M_{SO4}}}{\frac{E_{SO2}}{M_{SO2}} + \frac{E_{SO4}}{M_{SO4}}}$$

Where M_{SO2} and M_{SO4} are the molar masses of SO_2 and SO_4, respectively. By performing a comparison of the values of the monthly mean f_{ox} as calculated above with the values of f_{ox} calculated offline, we can determine that the non-linearities do result in quantitative differences. We therefore restrict ourselves to the weaker claim that the non-linearities do not qualitatively affect the analysis. It is unfortunately not possible to similarly calculate the monthly mean values of the other P6 outputs based on the available GEOS-Chem-TOMAS output, but we would expect that the non-linearities would result in quantitative differences in the values of those outputs as well.

P 21491

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?

The sensitivity of N_{new} to CS is between -1 and -2 % $\%^{-1}$ over most of the domain, but it is more positive than -1 % $\%^{-1}$ over small sections of inland Antarctica, and it is move negative than -2 % $\%^{-1}$ over polluted regions, such as the eastern United States, Europe, India, and eastern China. We have added the following sentence to the description of Fig. 5 to highlight this heterogeneity:

"A 1 % increase in CS yields a decrease in the predicted value of N_{new} of between 1 % and 2 % over most locations, and greater decreases over polluted regions such as the eastern United States, Europe, India and China (Fig. 5)."

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."

We have revised the sentences to the following:
"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 changes in sulphate formation and growth (at both the grid-resolved and sub-grid scales) due to increased $bgSO_2$ and $bgNO_x$ 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?

Over eastern China, the absolute number of sub-grid sulphate particles formed is less in the P6_hiSO2 simulation than in the P6_nXSOA_Napa simulation. Large increases in CS due to the additional SO_2 emissions suppress new-particle formation at the sub-grid scale, and the number of new particles per kg SO_2 decreases by more than 40% over a small area in eastern China, as shown in Fig. 8b. Even after accounting for the 50% increase in SO_2 emissions, this yields a net decrease in the absolute number of

sub-grid sulphate particles formed over this region, as shown in Fig. 9a. We have rephrased this paragraph to emphasize that Fig. 9 shows a relative difference in the absolute number of sub-grid sulphate particles formed.

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.

We have rephrased this sentence to the following:

"When using previous treatments of sub-grid sulphate, including an additional 100 Tg yr^{-1} of SOA led to an increase in the N80 attributable to sub-grid sulphate particles. This increase was due to an enhancement in condensational growth of the sub-grid sulphate particles."

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.

The following footnotes have been added to the table:

"AS3 - 3% of SO_2 emitted as sub-grid sulphate, 15% of sub-grid sulphate emitted into nucleation mode and remaining sub-grid sulphate emitted into Aitken mode.
LY5 - 5% of SO_2 emitted as sub-grid sulphate, 5% of sub-grid sulphate emitted into nucleation mode and remaining sub-grid sulphate condensed onto pre-existing aerosol.
P6 - fraction of SO_2 emitted as sub-grid sulphate, number and size of sub-grid sulphate particles dynamically predicted by P6 parameterization, remaining sub-grid sulphate condensed onto pre-existing aerosol."

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, nXSOA_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.

The table has been re-ordered and horizontal lines have been inserted.

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.

As shown in Fig. 8f and 8g, the number and size of sub-grid sulphate particles emitted decreased in the P6_hiNOx simulation from the P6_nXSOA_Napa simulation. The increases in particle number in this simulation at all sizes are therefore very likely to be due to increases in grid-resolved nucleation and condensation growth.

We would expect that the increased SO_2 emissions in simulation P6_hiSO2 would result in greater

H_2SO_4 concentrations globally, and therefore increased grid-resolved nucleation rates. We expect that the decreases in N3 and N10 in this simulation are due to enhanced coagulation, but not suppressed nucleation.

As shown in Fig. 9b, the number of sub-grid sulphate particles emitted was less over polluted regions in P6_hiboth than P6_nXSOA_Napa. Many of these regions are co-located with regions of increased N80 as shown in Fig. 7c. We therefore expect that the increases in N80 are not due to enhanced sub-grid sulphate emissions, but instead from enhanced condensation growth.

We have added the following to the caption:

"Decreases in N3 for the P6_hiSO2 and P6_hiboth cases are due primarily to enchanced coagulation. Increases in N40 and N80 for P6_hiSO2 and P6_hiboth are due primarily to enhanced condensational growth. Increases at all sizes for the P6_hiNOx case are due to enchanced grid-resolved nucleation and growth."

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?

We have increased the font size of the column and row titles.

The P6 parameterization generates a single lognormal mode with a constant geometric standard deviation of 1.4 for sub-grid sulphate particles. For constant N_{new}, decreasing D_m will shift the median diameter of the lognormal mode to a smaller size, resulting in both more smaller particles and fewer large particles, but it will not change the total number of particles emitted. In the specific case of the P6_hiNOx simulation shown in Fig. 8 (f), the value of N_{new} has also decreased, so both the total number of particles emitted and the median diameter of those particles decreased in that case. Both of these effects would decrease the number of large particles emitted, but it is not clear whether the number of particles smaller than the original median diameter has increased or decreased for any particular location.

Referee #2:

The paper discusses the implementation of the Predicting Particles Produced in Power-Plant Plumes (P6) parameterisation, described previously by Stevens & Pierce in 2013, within the GEOS-Chem-TOMAS global model.

This is a valuable contribution in terms of recognising the importance of sub-grid scale processes in accurately determining quantities that are important on the global scale. However, the authors are aware that at this stage "the differences in annually averaged aerosol size distributions due to the treatment of sub-grid sulphate at the measurement sites examined here are too small to unambiguously establish P6 as providing better agreement with observations".

The paper is well written, interesting and certainly within the scope of ACP; I would recommend publication, once the below (very minor and mainly technical) issues are addressed.

Specific / Minor Comments:

p 21476, line 5: "Currently..", I see what you mean but this sentence is confusing, I would suggest rewording.

We have rephrased the sentence to the following:

"Currently, the resolutions of regional- and global-scale models are typically at least tens or hundreds of kilometres, respectively. These models are therefore unable to accurately resolve the formation and growth of aerosols within these plumes using grid-box averages for chemical concentrations, aerosol concentrations, and meteorological values."

p 21477, line 1 (and line 3): this is a slightly ambiguous way of describing the ranges, do you mean increased by between 23 and 53% (otherwise this could be misinterpreted as increased by 23% to become 53%; that wouldn't make sense here but this is a point that is applicable to all quoted ranges)

We have rephrased the sentence to the following:

"Additionally, they found that CCN(0.2%) increased by either 23% or 53% averaged over the global boundary layer, and that the aerosol indirect effect radiative forcing increased by either 11% or 31% (depending on the grid-resolved nucleation scheme used in the boundary layer)."

p 21477, line 27: what do you mean when you say "used before"? In a previous study of theirs, or others? Or the range you discussed earlier?

Here we refer to the range of values used in the previous studies we cite in the preceding paragraphs. We have rephrased the sentence to the following for clarity:

"Based on the results of Stevens et al. (2012), the range of possible for the diameter of sub-grid-sulphate particles values used in Lee et al. (2013) was reduced to a smaller range than the full range of sub-grid-sulphate assumptions used in the studies cited in the preceding paragraphs."

p 21481, lines 13 – 18: very long sentence, I'd consider splitting this up for readability – in any case, replace "with" with "which" on line 17

We have rephrased the sentence to the following:

"This missing oxidation pathway would lead to an underestimation of the fraction of SO_2 oxidized on the sub-grid scale. However, little new-particle formation would be predicted under cloudy conditions because of the suppression of sunlight, which in turn would lead to lower OH concentrations and lower H_2SO_4 concentrations (Stevens et al., 2012). We therefore do not expect this missing oxidation pathway to strongly affect predictions of aerosol number."

p 21481, lines 20 - 23: You might want to rephrase this slightly, unless I have misunderstood – if "the SAM-TOMAS model has been shown to predict well the number and size of aerosol formed in coal-fires power plant plumes" then why is P6 needed?

The SAM-TOMAS model requires several hours of computation time to simulate the plume from a single power-plant to a distance to 50 km. It is therefore not suitable for implementation into a regional- or global-scale model. We have added additional description of the SAM-TOMAS model in order to clarify this:

“The P6 parameterization is based upon the results of the System for Atmospheric Modelling (SAM) (Khairoutdinov and Randall, 2003) with the TOMAS microphysics module described above. The SAM-TOMAS model is a complex Large-Eddy Simulation/Cloud Resolving Model capable of resolutions between tens of metres and hundreds of kilometres and domains between tens and hundreds of kilometres.”

p 21484, line 3 – 16: add somewhere here the size of particle being produced at these new particle formation rates

We have added the following sentence to the end of the paragraph:

“All nucleation schemes used in this study predict the formation rate of 1 nm particles. Aerosol growth and coagulational loss below 3 nm is approximated by the parameterization of Kerminen and Kulmala, (2002).”

p 21484, line 28: what do you mean by “actual SO2 emissions”? maybe just remove “actual” from this sentence, or clarify that you mean the global total, in contrast to the decreasing emissions from power plants in the US

Here, we intend to emphasize that our high-emissions simulations should not be interpreted as historical simulations. We have rephrased this sentence to the following:

“We note that real-world SO_2 emissions were not greater in the past for all locations globally, and the SO_2 emissions in this simulation are therefore not meant to represent any previous year but rather a general sensitivity to these emissions.”

p 21488, line 10: you could add here that although these previous studies suggest the formation of anthropogenically controlled SOA, the mechanism for this is not known, i.e., it’s not just that you didn’t include it in P6

We have rephrased the sentence to the following:

“These processes would compensate somewhat for the suppression of new-particle formation by the enhanced background condensation sink, but are not currently accounted for by the P6 parameterization because the mechanism(s) for the formation of this anthropogenically controlled SOA remain poorly understood.”

Additional citations:

Seinfeld, J. H. and Pandis, S. N.: Atmospheric Chemistry and Physics: From Air Pollution to Climate Change, 2nd ed., John Wiley and Sons, Inc., Hoboken, New Jersey., 2006.

Kerminen, V.-M. and Kulmala, M.: Analytical formulae connecting the “real” and the “apparent” nucleation rate and the nuclei number concentration for atmospheric nucleation events, J. Aerosol Sci., 33(4), 609–622, doi:10.1016/S0021-8502(01)00194-X, 2002.