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Interactive comment on "Comment on "Reduced efficacy of marine cloud brightening geoengineering due to in-plume aerosol coagulation: parameterization and global implications"" *by* S. Anand and Y. S. Mayya

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Comments from Dr J R Pierce (Referee 2)

I shared my general thoughts on the comment manuscript in the pre-ACPD review, and I'll repeat them in this paragraph. In this comment manuscript, the authors show an alternative method to solving coagulation in ship plumes to that of Stuart et al. (2013) (S13), a project that I supervised. We apologize to the authors of the comment that we were not aware of their 2011 work that provided a general framework to coagulation

in plumes – not only to ship plumes but plumes in general. Had we been aware of their work at the time, we may have used their framework as the basis for our work. Thus, I have no problem with the authors publishing a formal comment on our 2013 paper showing an alternative method. The particular advantage I see of their work is the more general applicability of their work outside of the range of input values tested in S13 (I think the comment authors can emphasize this advantage of their work in their revised version). In the originally submitted (pre-ACPD) version of the comment manuscript, there were several incorrect claims about the S13 work that have mostly been corrected; however one in correct claim remains (the emphasis on Turco and Yu's work) and another incorrect claim has been introduced (claiming that our work is a puff formulation) into this version. Once these incorrect claims have been fixed and several other minor comments have been addressed, this comment should be published in ACP. Incorrect claims (page/line numbers of the claims are pointed out in my specific comments):

1. Plume vs. puff models:

A lot of text regarding differences between puff and plume models has been introduced to this version of the manuscript (text that was absent from the original submission to ACPD). The comment manuscript authors claim that the work on S13 is based on puff assumptions rather than plume assumptions. This is incorrect as all of the work in S13 assumes plumes not puffs. Puffs expand by diffusion in the direction of the mean wind. This diffusion in the direction of the mean wind lowers particle concentrations in the puff (relative to a plume) and reduces coagulation rates (relative to a plume). We consider a Lagrangian slice of plumes that have no diffusion in the mean wind direction (or specifically, this assumes that gross diffusion in from an adjacent slice equals gross diffusion out to the adjacent slice creating no net diffusion between adjacent slices). Thus, issues of "inter-puff coagulation effects" noted in the comment manuscript are accounted for by our "no net diffusion in the direction of the mean wind" assumption of our slices, and the similarity of the results between the S13 and Anand and Mayya

(2011) (AM11) methods is not entirely surprising to me.

2. Numerical modeling vs. Turco and Yu:

Similar to the originally submitted (pre-ACPD) comment manuscript, the comment authors emphasize the use of Turco and Yu (1997) in S13 rather than the multi-slice, multi-size-bin numerical model that actually generated the data in S13. Turco and Yu (1997) was used in S13 to arrive at the sigmoidal equation for our parameterization (eqn. 5 in S13) and gain insight into the physics behind an analytical solution. We could have just as easily used Eqn. 47 of AM11 (same as Eqn. 2 of the comment manuscript) – which is also a sigmoid – to arrive at Eqn. 5 in Stuart et al. (2013) and gain this physical insight. (Again, we weren't aware of AM11 at the time of writing the paper, and we apologize.) Note that Turco and Yu also assume a plume not a puff.

3. Related to both of these points, there is mention that we assume uniform mixing. This is also incorrect since the numerical model has 10 radial shells to simulate the concentration gradients between the core and outside of the plume (see Fig. 1 in S13 for a schematic of this).

Author's Response to the general comments (1 to 3)

We thank the referee for his critical comments, especially his point about sealing axial diffusion to eliminate inter-puff coagulations (which we missed somehow as it was not clearly stated so in the Stuart et al. paper). In view of this, we thoroughly revised our manuscript to ensure that their work is not incorrectly represented. Thus, we have removed referring to their work as a "puff based, uniformly mixed model" and also the detailed discussions on puff versus plume model. However, it is important to note that the approach of Stuart et al. is based on fitting multiple, independent, exponents to the variables of the problem as opposed to our approach which involves a single exponent fit to a similarity variable that combines the relevant variables. To bring out this point, we have now introduced the terminology of single exponent versus multi-exponent, as the basic differences in the two approaches. Also ours is not strictly a sigmoidal

solution and predicts different power-law behaviour for limiting cases of the sources strength. These points have been inserted in the abstract and at appropriate places in the revised manuscript.

Author's changes in manuscript

Changes in manuscript are mentioned at the appropriate specific comments below.

Specific comments:

1. P23798 L4: "While the Stuart et al.'s approach is based on the solutions to the coagulation problem in a uniformly mixed expanding puff model...". There is no *puff* model used in Stuart et al. (2013) (S13), nor do we assume that it is uniformly mixed. See the general comment above.

2. P23798 L7: "We discuss the conceptual differences between the survival fraction estimates from standing plume models as opposed to that from puff models." Again, no puffs.

Author's Response to comment 1 and 2:

The restriction of axial diffusion was not clear in the manuscript of Stuart et al. and hence the comparisons between puff and plume models were made in the earlier version. In the present revised version, we have taken note of the detailed explanation (on sealing the axial diffusion) and other comments of the referee and modified the text accordingly.

Author's changes in manuscript

While the Stuart et al.'s approach is based on the solutions to the coagulation problem in an expanding plume model, the diffusion based approach solves the diffusion-coagulation equation for a steady-state standing plume to arrive at the survival fraction correlations. We discuss the differences in the functional forms of the survival fraction expressions obtained in the two approaches and compare the results for the case

studies presented in Stuart et al. (2013) involving different particle emission rates and atmospheric stability categories.

3. P23798 L9: "The two models predict different functional forms for dependencies of the survival fraction on source and atmospheric related parameters." Both models arrive at sigmoid functions, although the terms of the sigmoid are different.

Author's Response:

The solutions from the two models have different functional forms; Stuart et al. have sigmoidal form $1/(1 + x^n)$ and we have the form $1/(1 + x)^n$, which is not exactly sigmoidal since it will not have a shoulder for $n >> 1$. Although the results are same in the present case study, the limiting results show some differences between them. These points are now discussed and added in the main text (in the Results and Discussion section as shown below).

Author's changes in manuscript

However, it must be reiterated that the two models are based on different formulational premises and predict different forms of the survival fraction on source related and turbulence related parameters. For example, in the limit of low particle emission rate $(P \to 0)$, Eq.(2) of our model predicts that the depleted/consumed particle fraction $(1 - F) \to \mu \propto P$ whereas Eq.(5) of S13 predicts a power-law dependence of the form $(1 - F) \propto P^c$, with *c* ranging from 0.51-0.76. On the other hand, in the limit of large emission rates, both the models predict a power-law decline of *F* with respect to *P*, with similar, if not identical, powers.

4. P23798 L15: "The diffusion based models have the inherent capability to generate similarity parameters with inbuilt exponents and hence avoid the parameterization exercise." But eqn. 47 of AM11 (same as eqn 2 in the comment manuscript) is a fit to the numerical solution. From AM11, "Since the quantity of crucial importance is the asymptotic survival fraction which will be a function of only one parameter, we have

numerically computed this quantity over a wide range of μ. For practical applicability, we fitted a function to these values, having a mathematical form analogous to that rigorously obtained in the puff case to maintain consistency between the formulae. The fitted formula for the plume model is Eqn 47." Thus, the AM11 method is not free of parameterization of exponents.

Author's Response:

We agree that the survival fraction formula from the diffusion based models also involves a "fit". However, this fit is a single exponent fit to a similarity variable μ. The choice of the variable μ is not arbitrary, but stems from a scaling analysis of the original equation. Similarly, the choice of the fitting function comes from comparison with asymptotic analytical results. So, the difference between the two approaches may justifiably be termed as single exponent versus multi-exponent parametrization. The text in the abstract is now modified to make this point clear.

Author's changes in manuscript

The diffusion based expression involves a single exponent fit to a theoretically generated similarity variable combining the parameters of the problem with in-built exponents and hence avoids the multi-exponent parameterization exercise.

5. P23798 L17: "However, their limitation lies in the choice of a representative value for the coagulation coefficient in an evolving aerosol system, which has been addressed in a more satisfactory manner by the parameterization method." I was also confused when I read this in the abstract, but again I figured it out when reading the same sentence when it appeared later in the body of the comment manuscript. Please make it clear that "their" is referring to diffusion based models (e.g. AM11) and "the parameterization method" is S13. When I first read the sentence, I thought that "their" was referring to S13.

Author's Response:

The point is taken care, and the text is modified accordingly.

Author's changes in manuscript

Abstract: However, in the diffusion model, the choice of a representative value for the coagulation coefficient is more prescriptive than rigorous, which has been addressed in a more satisfactory manner by the parameterization method.

Manuscript text: Since a significant part of the coagulation effect is expected to occur near the source region, where the particle concentration will be the highest, we use the value of the effective coagulation coefficient (K_c) of the initial aerosol spectrum. This may be viewed as a model prescription which may not be entirely satisfactory for an evolving aerosol spectrum.

6. P23799 L1: "The authors base their work on the model proposed earlier by Turco and Yu (1997) to estimate the fraction of particles surviving coagulation (survival fraction) within a dispersing air packet (volume element)." Our work is not based on Turco and Yu (1997), we simply used Turco and Yu's solution of coagulation in a plume (not *plume*, not *puff*) to arrive at the sigmoidal equation for our parameterization (eqn. 5 in S13) and gain insight into the physics behind an analytical solution. See my general comment above. Differences between the S13 parameterization and the numbers calculated in the comment manuscript would mostly be due to differences between the numerical model in S13 (or more correctly, the fit of the numerical model) and AM11. Please remove the focus on Turco and Yu in this introductory paragraph.

7. P23799 L3: "The Turco-Yu model treats this problem within the framework of a solving the coagulation equation in a uniformly mixed aerosol puff volume which is expanding at a prescribed rate in time. The simplifying feature of this model is that it replaces the gradient driven nature of the dispersion process by a purely time dependent term leading to an analytically tractable solution to the survival fraction. It is implicitly assumed that the survival fraction estimated in an expanding puff (Lagrangian framework) is applicable to standing plumes (Eulerian framework). S13 further extend

this approach by considering several strata of different concentration domains in the plume and relating the survival fraction to five atmospheric dispersion and source related parameters." As per the previous comment, this text is generally irrelevant. The numerical model in S13 created the data, not Turco and Yu. Also, neither the numerical model nor Turco and Yu uses a puff assumption.

Author's Response to comment no. 6 and 7:

As suggested by the reviewer, we have now removed the discussions about Turco and Yu model in the revised manuscript.

Author's changes in manuscript

A parameterization scheme is provided by Stuart et al. (2013) (hereafter, S13) to assess the loss of particle number concentration by coagulation in plumes for cloud-resolving and global models. The authors numerically solve the coagulation problem in a dispersing plume, and employ a multi-exponent parameterization scheme to obtain a semi-empirical equation by fitting their multi-shelled Gaussian plume model to five atmospheric dispersion and source related parameters. The fitted formula is then used to estimate the fraction of particles surviving coagulation (survival fraction) within a dispersing plume volume. The choice of the functional form of empirical equation in S13 is based on the survival fraction formula provided earlier by Turco and Yu (1997) within the framework of solving the coagulation equation in a volume which is expanding at a prescribed rate in time. The simplifying feature of the Turco and Yu model (1997) is that it replaces the gradient driven nature of the dispersion process by a purely time dependent term leading to an analytically tractable solution to the survival fraction.

8. P23799 L13: "In contrast to the uniformly mixed, expanding puff model..." The data in S13 was created with a model that was neither uniformly mixed (10 radial shells in the numerical model) nor a puff model.

Author's Response:

We agree to this comment, and deleted reference to 'uniform mixing' and 'puff'.

Author's changes in manuscript

As an alternative to the above approach, Anand and Mayya (2009, 2011) have developed a formalism based on solving the coagulation-diffusion equation for estimating the survival fraction of aerosols in dispersing puffs and plumes.

9. P23799 L28: "It may be recalled that (Seinfeld and Pandis, 2006) while a plume can be treated exactly as a limiting case of a train of puffs for nonreactive dispersions, nonlinear reaction processes such as coagulation do not yield identical results for the survival fraction in the two cases. This is because, the inter-puff coagulation effects, which play a dominant role in the asymptotic survival of particles in a plume are neglected in puff calculations." This discussion is moot because we don't consider a train of puffs (which diffuse in the mean wind direction). Rather we consider a slice that has no net diffusion in the mean wind direction (which is appropriate when the mean wind speed greatly exceeds turbulent diffusion in the mean wind direction). Inter-slice coagulation effects are considered in our method by assuming no net diffusion in the direction of the mean wind (this assumption is that the gross diffusion between adjacent slices balance each other to create no net diffusion).

Author's Response:

We have addressed this point at various places previously in this reply. We again confirm that the usage of puff is now completely removed from the manuscript.

Author's changes in manuscript

The text mentioned in this comment (no. 9) is deleted in the present revised manuscript.

10. P23800 L27: "On the other hand, the present model captures coagulation characteristics through a single parameter Kc, whereas S13, use polydispersity index (sigma) and particle diameter (Dp) separately to account for coagulation.". The fit includes ini-

tial sigma and the initial median diameter; however, the coagulation in numerical model in S13 is determined throughout the 10 shells and 100 size sections. As the size distribution evolves the mean coagulation coefficient of the system evolves (the sigma and mean diameter will change with time). Thus, the evolution of the coagulation coefficient in the numerical model depends on all of the inputs; i.e. the emissions rate, wind speed and stability all affect how the size distribution changes with time and thus how the coagulation coefficient changes with time. This evolution of the coagulation coefficient is captured implicitly in the fits in S13. How should the user of AM11 go about determining which Kc to use for a given sigma and median Dp? Advice for this?

Author's Response:

Although one can use time-dependent coagulation coefficient (Kc(t)) to account for the evolving size spectra with respect to time, we have used effective constant coagulation coefficient (Kc) of the initial size distribution in the present calculations. This limitation is clearly mentioned in the present manuscript as shown below. Also, one can note that the variation of the mean dry diameter and sigma of the size distribution leads to marginal change in the survival fraction (Fig.5 of S13).

Author's changes in manuscript

On the other hand, the present model captures coagulation characteristics through a single parameter Kc whereas S13, use polydispersity index (σ) and particle diameter (Dp) separately to account for coagulation. Since a significant part of the coagulation effect is expected to occur near the source region, where the particle concentration will be the highest, we use the value of the effective coagulation coefficient (Kc) of the initial aerosol spectrum. This may be viewed as a model prescription which may not be entirely satisfactory for an evolving aerosol spectrum.

11. P23802 L8: "Seen from this perspective, the diffusion based models have the inherent capability to generate similarity parameters with inbuilt exponents and hence avoid the parameterization exercise." See the comment I made for the same sentence

that appeared in the abstract.

Author's Response:

In the present revised version, the text is modified to show the difference between single- and multiple- exponent fitting. Please see the author's response and author's change in the manuscript for the comment no.: 4 in addition to this reply.

Author's changes in manuscript

Seen from this perspective, the diffusion based model has the inherent capability to generate a similarity variable with inbuilt exponents for the parameters and hence avoids the multi-exponent parameterization exercise.

12. P23802 L10: "However, their limitation lies in the choice of a representative value for the coagulation coefficient in an evolving aerosol system, which has been addressed in a more satisfactory manner by the parameterization method." Again, please make it clear that "their" is referring to diffusion based models (e.g. AM11) and "the parameterization method" is S13.

Author's Response:

Yes, we ensured that the ambiguity of 'their' is now removed.

Author's changes in manuscript

However, the limitation of the diffusion model is that it does not provide a rigorous framework for the choice of a representative value for the coagulation coefficient in an evolving aerosol system, which has been addressed in a more satisfactory manner by the parameterization method (S13).