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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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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

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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.

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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.

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).

Specific comments:

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.

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.

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.

P23798 L15: “The diffusion based models have the inherent capability to generate

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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.

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.

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.

P23799 L3: “The Turco-Yu model treats this problem within the framework of a solving

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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.

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.

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).

P23800 L27: “On the other hand, the present model captures coagulation characteristics through a single parameter K_c , whereas S13, use polydispersity index (σ)

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and particle diameter (D_p) separately to account for coagulation.". The fit includes initial 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 K_c to use for a given sigma and median D_p ? Advice for this?

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.

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.

Interactive comment on Atmos. Chem. Phys. Discuss., 14, 23797, 2014.

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