Comment on "Reduced efficacy of marine cloud brightening geoengineering due to in-plume aerosol coagulation: parameterization and global implications"

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Abstract

We examine the parametrized model of Stuart et al. (2013) *vis-a-vis* a diffusion based model proposed by us earlier (Anand & Mayya, 2011) to estimate the fraction of aerosol particles surviving coagulation in a dispersing plume. 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. There appear to be a better agreement between the two models at higher survival fractions as compared to lower survival fractions; on the whole, the two models agree with each other within a difference of 10%. 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. It also possesses a wider range of applicability in respect of the source and atmospheric parameters as compared to that based on parametrization. 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. The present comparative exercise, although limited in scope, confirms the importance of aerosol microphysical processes envisaged by Stuart et al. for cloud brightening applications. In a larger context, it seems to suggest that either of the two forms of expressions might be suitable for incorporation into global/regional scale air pollution models for predicting the contribution of localized sources to the particle number loading in the atmosphere.

1 Introduction

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.

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. In their 2011 work, they specifically addressed the issue of particle number survival fraction in a standing plume, maintained by a steady emission source, by combining turbulent diffusion and advection with coagulation through an equation of the form

$$v_w \frac{\partial N}{\partial x} = \frac{v_w}{4}\frac{d\sigma^2}{dx}\left[\frac{1}{r}\frac{\partial}{\partial r}\left(r\frac{\partial N}{\partial r}\right)\right] - \frac{K_c}{2}N^2. \quad (1)$$

In Eq.(1), N is the particle number concentration, v_w is the wind speed, σ is the plume width (expressed through a spatially varying turbulent diffusion coefficient), x, r are the down-wind and the cross wind coordinates respectively and K_c is an effective coagulation coefficient, taken as size independent constant. The source emission rate provides the flux matching condition at x=0. Basically, this model provides a mechanistic basis for dispersion to estimate the survival fraction in an Eulerian framework; further, it directly solves Eq.(1) to obtain the number concentration profile in a standing plume.

Given the steady-state nature of the above approach, the survival fraction is evaluated rather differently as compared to time dependent expansion problems. It is defined as the ratio of the flux of particles integrated over the entire cross section at a down-stream distance x, to that emitted in the source domain, in the limit, $x \to \infty$. A scaling analysis of Eq.(1) showed that the survival fraction (F) is a unique function of a similarity variable μ (see below) that combines all the parameters of the problem with inbuilt exponents. Further, a limiting analysis indicated that F should possess a functional form of the type $(1+\mu/\nu)^{-\nu}$, where, ν is an exponent to be determined. Upon combining these results with numerical solution of Eq.(1) for evaluating ν through a single exponent fit, the survival fraction was then represented in terms of the variable μ, in the following form:

$$F = \frac{1}{(1+1.32\mu)^{0.76}} \tag{2}$$

where, $$\mu = \frac{K_c P}{6\sqrt{3} v_w (2R_s)^{4/3} (C\varepsilon)^{1/3}}, \tag{3}$$

K_c is the effective coagulation coefficient, P is the number emission rate, v_w is the wind velocity, R_s is the emission stack radius (plume radius at the source of emission), C is a constant (0.8), and ε is the turbulent kinetic energy dissipation rate. As in the case of S13, the present result also involves five parameters all combined in a single variable μ. However, there are subtle differences: the present model involves two parameters to describe atmospheric conditions (v_w and ε) whereas S13 account for this through a single parameter (v_w). On the other hand, the present model captures coagulation characteristics through a single parameter K_c whereas S13, use polydispersity index (σ) and particle diameter (D_p) 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 (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. An important point about the present model is that, the survival fraction formula (Eq.(2)) can be applied beyond the fitting range of input values tested in S13, and it provides a general framework to coagulation of aerosols in plumes (e.g. forest fires, volcanic emissions, etc.).

The quantity ε, the turbulent kinetic energy dissipation rate (Table 1), may be estimated for different atmospheric stability classes through the well-known relationships of atmospheric boundary layer theory (Hans *et al*., 2000). In the Table 1, $x = \left[1 - 15\frac{z}{L}\right]^{1/4}$, L is Monin-Obukhov length, u_* is the friction velocity (Stull, 1988), z is the height of release, z_0 is the roughness length, k is the van Karman constant (0.4), and u is the wind velocity. The Monin-Obukhov length (L) is obtained using a fitting expression (Seinfeld and Pandis, 2006) for various stability categories and roughness length. The L values obtained corresponding to a z_0 of 0.02 m (oceanic surface) are, -11.6 for unstable, ∞ for neutral, and 10.4 for stable categories, and these are used in the present study.

2 Results and Discussion

We now compare the estimates of the survival fractions from these two models using the case studies described in S13 and the values presented in their Table 1 for the wind speed, particle emission rates and stack radius. In the present calculations, the atmospheric stability classes A, B, C have been combined into a single (unstable) category, and the classes E and F have been combined into one "stable" category. The category D (neutral) has been retained as such.

The results of the survival fractions obtained with the two approaches are tabulated in Table 2. The survival fraction values obtained for "Minimum", "Base", and "Maximum" cases (Table 2) correspond respectively to the minimum, base, and maximum of all the five parameters mentioned in the Table 1 of S13. The number survival fraction estimated from the present model varies from 0.36 to 0.62, thereby confirming the important role of aerosol microphysical processes as envisaged by S13, in significantly altering the source to receptor transfer of particles for cloud brightening applications.

Excepting in the E/F category for the "maximum" case, the survival fraction estimates from the two approaches for all other cases are rather close to each other. Both the models seem to predict similar trends: survival fractions are lower for increasing emission rate and/or increasing atmospheric stability. There appear to be better agreement between the two models at higher survival fractions and relatively poorer agreement at lower survival fractions. On the whole, it is still remarkable that both the models are close to each other within 10%.

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 $(P \rightarrow 0)$, Eq.(2) of our model predicts that the depleted/consumed particle fraction $(1 - F) \rightarrow \mu \propto P$ 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. It will be rewarding to explore the implications of these approaches in the general context of atmospheric aerosols for estimating the contribution of various anthropogenic sources to background particles. 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. 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). Notwithstanding these issues, the present numerical comparisons, although limited in scope, seems to suggest that either of the two forms of expressions might be suitable for incorporation into global/regional scale air pollution models for predicting the contribution of localized sources to the particle number loading in the atmosphere.

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Table 1. Friction velocity (u_*) and turbulent kinetic energy dissipation rate (ε) for various atmospheric stability categories. (See text for definition of quantities x, L, z_0)

Stability category	Friction velocity (u_*), m s^{-1}	TKE Dissipation rate (ε), m^2 s^{-3}
Unstable	$ku\left[ln\left(\frac{z}{z_0}\right)-2ln\left(\frac{1+x}{2}\right)-ln\left(\frac{1+x^2}{2}\right)+2tan^{-1}x-\frac{\pi}{2}\right]^{-1}$	$\frac{u_*^3}{kz}\left(1+0.5\left\|\frac{z}{L}\right\|^{2/3}\right)^{3/2}$
Neutral	$ku\left[ln\left(\frac{z}{z_0}\right)\right]^{-1}$	$\frac{u_*^3}{kz}\left(1.24+4.3\frac{z}{L}\right)\left(1-0.85\frac{z}{h}\right)^{3/2}$
Stable	$ku\left[ln\left(\frac{z}{z_0}\right)+\frac{4.7(z-z_0)}{L}\right]^{-1}$	same as above

Table 2. Number survival fraction in a plume obtained using the two models.

<table>
<tr><th rowspan="3">Stability category</th><th colspan="6">Number survival fraction</th></tr>
<tr><th colspan="2">Minimum</th><th colspan="2">Base</th><th colspan="2">Maximum</th></tr>
<tr><th>Eq.(5) of S13</th><th>Eq.(2)[a]</th><th>Eq.(5) S13</th><th>Eq.(2)[a]</th><th>Eq.(5) of S13</th><th>Eq.(2)[a]</th></tr>
<tr><td>A</td><td>0.629</td><td rowspan="3">0.621</td><td>0.562</td><td rowspan="3">0.544</td><td>0.515</td><td rowspan="3">0.495</td></tr>
<tr><td>B</td><td>0.626</td><td>0.549</td><td>0.497</td></tr>
<tr><td>C</td><td>0.589</td><td>0.492</td><td>0.429</td></tr>
<tr><td>D</td><td>0.547</td><td>0.507</td><td>0.436</td><td>0.43</td><td>0.368</td><td>0.384</td></tr>
<tr><td>E</td><td>0.505</td><td rowspan="2">0.481</td><td>0.379</td><td rowspan="2">0.405</td><td>0.303</td><td rowspan="2">0.361</td></tr>
<tr><td>F</td><td>0.404</td><td>0.266</td><td>0.191</td></tr>
</table>

[a] Eq.(47) of Anand and Mayya (2011) is reproduced as Eq.(2) in the present work.