Atmos. Chem. Phys. Discuss., 14, 23797–23805, 2014
www.atmos-chem-phys-discuss.net/14/23797/2014/
doi:10.5194/acpd-14-23797-2014

Atmospheric Chemistry and Physics
Discussions
Open Access

This discussion paper is/has been under review for the journal Atmospheric Chemistry and Physics (ACP). Please refer to the corresponding final paper in ACP if available.

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

S. Anand[1] **and Y. S. Mayya**[2]

[1]Health Physics Division, Bhabha Atomic Research Centre, Trombay, Mumbai – 400 085, India
[2]Department of Chemical Engineering, Indian Institute of Technology-Bombay (IIT-B), Powai, Mumbai – 400 076, India

Received: 22 July 2014 – Accepted: 1 September 2014 – Published: 15 September 2014

Correspondence to: Y. S. Mayya (ysmayya@iitb.ac.in)

Published by Copernicus Publications on behalf of the European Geosciences Union.

Abstract

We examine the parametrized model of Stuart et al. (2013) vis-a-vis a diffusion based model proposed by us earlier (Anand and 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 a uniformly mixed expanding puff model, the diffusion based approach solves the diffusion-coagulation equation for a standing plume to arrive at the survival fraction correlations. We discuss the conceptual differences between the survival fraction estimates from standing plume models as opposed to that from puff models. The two models predict different functional forms for dependencies of the survival fraction on source and atmospheric related parameters. We compare the results for different case studies presented in Stuart et al. (2013) involving different particle emission rates and atmospheric stability categories. There appear to be 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 models have the inherent capability to generate similarity parameters with inbuilt exponents and hence avoid the parameterization exercise. 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. The present comparative exercise, 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.

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-

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resolving and global models. 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). The Turco–Yu model treats this problem within the framework of 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.

In contrast to the uniformly mixed, expanding puff model, Anand and Mayya (2009, 2011) have developed an alternative 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 plume maintained by a steady emission source by combining turbulent diffusion and advection with coagulation through an equation of the form

$$U\frac{\partial N}{\partial x} = \frac{U}{4}\frac{\mathrm{d}\sigma^2}{\mathrm{d}x}\left[\frac{1}{r}\frac{\partial}{\partial r}\left(r\frac{\partial N}{\partial r}\right)\right] - \frac{K}{2}N^2. \qquad (1)$$

In Eq. (1), N is the particle number concentration, U 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 and K 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; further, it allows one to treat the survival fraction within an Eulerian framework by directly considering a standing plume without having to consider the fate of particles in a train of individual expanding puffs. It may be recalled that (Seinfeld and

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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. Anand and Mayya (2011) (hereafter, AM11) demonstrated this effect explicitly for a hypothetical case of constant diffusion coefficient: within the rigorous standing plume model, the survival fraction of particles decreases monotonically to zero as one moves away along the plume in contradistinction to a spherical puff in which the ultimate survival fraction is non-zero finite. Thus for steady releases, the standing plume model offers a direct conceptual advantage in terms of rigor over expanding puff based models for handling coagulation effects.

The survival fraction for a standing plume model is evaluated 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, $x \rightarrow \infty$. By combining numerical solutions with the analytical results of an asymptotic theory, the survival fraction was then represented in terms of a single parameter μ, in the following form:

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

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

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

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characteristics through a single parameter K_c, whereas S13, use polydispersity index (σ) and particle diameter (D_p) separately to account for coagulation.

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 (Han et al., 2000). In the Table 1, $x = [1-15\frac{z}{L}]^{1/4}$, L is Monin–Obukhov length, u^* is the friction velocity (Stull, 1988), z is the height of release, z_0 is the roughness length, h is the height of atmospheric boundary layer, 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 to the minimum, base, and maximum of all the five parameters mentioned in the Table 1 of S13. 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 frac-

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tions 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 functional dependencies of the survival fraction on source related and turbulence related parameters. 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 models have the inherent capability to generate similarity parameters with inbuilt exponents and hence avoid the parameterization exercise. 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. On the whole, the present comparative exercise, 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 for various atmospheric stability categories. (See text for definition of quantities x, L, z_0.)

Stability category	Friction velocity (u^*), $\mathrm{m\,s^{-1}}$	TKE Dissipation rate (ε), $\mathrm{m^2\,s^{-3}}$
Unstable	$ku\left[\ln\left(\frac{z}{z_0}\right) - 2\ln\left(\frac{1+x}{2}\right) - \ln\left(\frac{1+x^2}{2}\right) + 2\tan^{-1}x - \frac{\pi}{2}\right]^{-1}$	$\frac{u_*^3}{kz}\left(1 + 0.5\left\|\frac{z}{L}\right\|^{2/3}\right)^{3/2}$
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

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

Stability category	Number survival fraction					
	Minimum		Base		Maximum	
	Eq. (5) of S13	Eq. (47)[a] of AM11	Eq. (5) S13	Eq. (47)[a] of AM11	Eq. (5) of S13	Eq. (47)[a] of AM11
A	0.629	0.621	0.562	0.544	0.515	0.495
B	0.626		0.549		0.497	
C	0.589		0.492		0.429	
D	0.547	0.507	0.436	0.43	0.368	0.384
E	0.505	0.481	0.379	0.405	0.303	0.361
F	0.404		0.266		0.191	

[a] Eq. (47) of AM11 is reproduced in this comment as Eq. (2).

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