

Interactive comment on “Review and uncertainty assessment of size-resolved scavenging coefficient formulations for snow scavenging of atmospheric aerosols” by L. Zhang et al.

Anonymous Referee #3

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General Comments:

This manuscript summarizes uncertainties in formulations of the impaction scavenging coefficients used to describe collisions between aerosols and snow. The authors review a number of parameterizations for collision efficiency, snow size distribution, snow fall velocity, and cross-sectional area, which are used to calculate scavenging coefficients, which describe aerosol-snow collisions and collection. The various parameterizations are compared and differences as large as three orders of magnitude are demonstrated. The parameterizations that bound the limits are applied to describe the time evolution of urban and marine aerosol number and mass concentrations, and

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the differences are investigated for two different snow intensities. The manuscript provides a useful summary of the uncertainties in aerosol impaction scavenging by snow and addresses an important scientific problem. The following concerns should be satisfactorily addressed prior to publication.

My major concern is related to the comparison between the theoretical and empirical scavenging coefficient formulations. These formulations should not be expected to agree with each other given that the latter are derived from field measurements, which include the additional effects of storm dynamics. Details are provided below in points 4 and 15. As well, certain of the assumptions made in the paper are not well justified, particularly, the suggested correction for the empirical parameterization (point 14 below). Additionally, portions of the discussion of the figures are rather cursory and should be further developed as indicated below (point 16).

Specific Comments:

1) Page 14826, line 26: In regard to terminology, this line indicates that the terms ice crystals and snow crystals can be used interchangeably. However, for consistency in the manuscript, it might helpful to select and use one of these terms throughout the discussion. Additionally, the manuscript should be clear about the size range of the collectors under consideration in this study, and focus the discussion on this size range. Page 14831, lines 16-18, discuss snow crystal habits and later in that paragraph uses the term ice crystals, yet at this point I am thinking that the study was focused on scavenging by precipitation size snow, not ice or snow crystals. The text often uses the term snow particles, please state the applicable size range that will be the focus of this study.

2) Page 14828, lines 6-10: Since collection efficiency is dimensionless, should the words ‘per unit time’ be added as follows: ‘normalized by the number of upstream particles of diameter d_p swept per unit time across an area. ...’?

3) Page 14833, line 14-15: Perhaps consider adding a reference related to the impor-

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tance of electric charges in the 0.1-1 μm size range e.g. Tingsley et al., 2000.

4) With respect to phoretic effects, Paramonov et al. 2011 found that relative humidity was the factor that correlated best with their scavenging coefficients derived from field measurements, whereas the previous discussion has dismissed these effects except for a limited aerosol size range. Could the discussion address this point? I think that this highlights the point that empirical scavenging coefficients derived from field measurements and theoretical scavenging coefficients are not the same thing. They cannot be compared for a given aerosol size with the expectation that they should be equal. Could the manuscript address this issue more explicitly? This relates back to earlier work by Flossman et al. (1991), Andronache et al. (2006) and Wang et al. (2011) and more recently by Qu  rel et al. (in press), which indicates that various dynamical factors related to storms influence the scavenging coefficients derived from field observations. This is unlike the scavenging coefficients derived from laboratory measurements and theoretical calculations.

5) Page 14836, lines 17-18: The formulas of Mitchell and Heymsfield (2005) and Murakami et al. (1985) were chosen for VD and E, respectively. Were the differences tested for any other combinations of VD and E parameterizations? Why were these particular ones chosen? Would the results differ for other combinations?

6) Page 14836, lines 20-21: The difference in the snow scavenging coefficients is noted to increase with increasing snow intensity. Can you comment on whether this increase is linear or not? This is difficult to determine with only two snowfall intensities shown. What would result for 1 mm h⁻¹?

7) Page 14839, line 27: Again only one particle size spectrum was considered for the sensitivity tests. Why was this spectrum chosen? How might the results change for different spectra?

8) Page 14840, line 21-22: The combined uncertainties are noted to be larger than for the individual parameters. Please indicate more explicitly if this is for a certain aerosol

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size range or related to a specific set of parameterizations.

9) Page 14840, lines 23-24: Consider stating explicitly the sizes or parameterizations that exhibit each of these behaviors i.e. the cancellation or enhancement of the uncertainties.

10) Figs 7 and 8: These figures are for snowfall intensities of 0.1 mm h⁻¹ and 1 mm h⁻¹. How would the results look for 10 mm h⁻¹?

11) Page 14840, lines 26-29: Consider making the discussion more quantitative. How much do the uncertainties increase with the increase in intensity?

12) Section 4.2: The Paramonov et al. (2011) empirical parameterization is dependent on relative humidity (RH). Please indicate the RH used for these calculations.

13) Page 14841, line 18-20: If I understand correctly, the Kyr   et al. (2009) parameterization is applicable for the snowfall intensity of 0.1 mm h⁻¹. This is one of the snowfall rates considered in this study. Perhaps consider including an indication how this compares to the Paramonov et al. (2011) parameterization and the given results here.

14) Page 14841, lines 26-28: Is this a valid assumption: that all snowfall intensities are equally likely to occur? The histogram in Fig. 3 of Paramonov et al. (2011) suggests that this is not the case and that snowfall intensities around 0.1 mm h⁻¹ were considerably more frequent than snowfalls of about 1 mm h⁻¹. That figure shows that almost 60% of the snowfalls had intensities of 0.2 mm h⁻¹ or less. Thus it is not clear to me that this discussion about a correction of the Paramonov et al. (2011) scavenging coefficients based on snowfall intensity is justified. Additionally, those authors found a low correlation between intensity and the scavenging coefficients derived from field measurements.

15) Section 4.2: This point relates to one of my major concerns with the paper in its present form. I think that the entire discussion related to the comparison between

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field measurements and theoretical calculations should be more carefully framed. It is not clear to me that empirically-estimated scavenging coefficients based on field measurements can be directly compared to theoretically calculated coefficients. This is somewhat of an apples and oranges comparison as also indicated in the references given in point 4 above. If the authors do decide to show this type of comparison, the text should be clearer that these coefficients are not expected to equate with each other. The empirically-estimated coefficients include the influence of storm dynamics and as such they might be appropriate to apply in a simple model which does not include any dynamical effects and thus used to describe the change in tracer concentrations. However, theoretically derived coefficients do not include any dynamical effects, and include only certain physical scavenging processes as the authors have indicated (Brownian motion, interception and impaction). Such theoretical coefficients are then suitable to be applied in more complex global models, which already include representations of the dynamical effects. In the existing framework of this study, empirical and theoretical coefficients cannot be expected to agree or to give similar results for the evolution of aerosol concentrations. This paper offers the opportunity to provide a demonstration/discussion of this point, if the presentation is carefully framed.

16) Section 4.3: The text here should provide a more complete discussion of Fig. 9. Please add an explicit discussion of the first row of panels of Fig. 9. Also, the figure shows both marine and urban scenarios, but there is no discussion in the text of the differences between these scenarios.

17) Page 14842, lines 25-26: The text states that the impacts of the differences in scavenging coefficient parameterizations are quantitatively different for number and mass. Please consider adding an explicit description of the main differences.

18) Page 14843, lines 15-17: By 'more aerosol particles', do you mean more in terms of number or mass or both?

19) Page 14844, lines 22-24: As indicated in point 14), I am not sure that this adjust-

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ment to the scavenging coefficients is well justified.

20) Page 14845, lines 26-28: The authors suggest development of a semi-empirical parameterization. How would this be different from previous parameterizations, such as those of Table 1. If this parameterization is based on field measurements, could this really be applied in 'any' size-distributed particulate-matter model i.e. how could this parameterization be equally applicable to models that do or do not already include storm dynamics?

21) Fig. 4: The differences between the lines, comparing between the panels, look quite similar and quite independent of snow particle shape. How might the figure appear if the Slinn parameterization was implemented? Would there be greater differences in the scavenging coefficients for certain shapes?

22) Fig. 8: How would the figure appear if you included 10 mm h⁻¹?

23) Abstract: Last sentence: Could the authors briefly mention the evidence provided by this study that supports this point?

24) Abstract: Line 17-19: Are the theoretical and empirically derived values expected to agree? I think this comparison (if it is made) needs to be more correctly framed since the latter includes storm dynamic effects not included in the former. Thus, we might not expect them to agree.

25) Abstract, line 20: Perhaps indicate the range of the scavenging coefficient differences that yields this factor of two aerosol concentration difference and specify whether you mean mass or number concentrations.

26) Abstract: Shape differences are not mentioned in the abstract, are the authors able to add anything in this regard from their study?

27) In general, I think the manuscript should acknowledge at some point that these uncertainties are quantified only for the parameterizations tested, which might not be an exhaustive list.

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28) Title: Perhaps consider adding the word 'impaction' before the words 'scavenging coefficient formulations' in the title.

Technical Corrections:

1) Fig. 2 caption: Change 10.0 m h⁻¹ to 10.0 mm h⁻¹

2) Figs. 5 and 6: Could the readability of the symbols to distinguish between lines of the same color be improved in this figure? Within a given color, would dashed and dotted lines be easier to see, rather than the triangles and circles on the lines?

3) Fig. 7 caption: Add '(as liquid water equivalent)' after the snowfall intensity.

4) Fig. 8 caption: Add 'with respect to Fig. 7' after 'note change in y-axis range'.

References:

Flossman, A.: The scavenging of two difference types of marine aerosol particles calculated using a two-dimensional detailed cloud model, *Tellus* 43B, 1991.

Quérel A., M. Monier, A. I. Flossmann, P. Lemaitre, and E. Porcheron: The importance of new collection efficiency values including the effect of rear capture for the below-cloud scavenging of aerosol particles, *Atmos.Res.*, (in press).

Tinsley, B. A., R. P. Rohrbaugh, M. Hei, and K.V. Beard: Effects of image charges on the scavenging of aerosol particles by cloud droplets and on droplet charging and possible ice nucleation processes, *JAS*, 57, 2000.

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