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Atmospheric Chemistry and Physics
Discussions

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

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We greatly appreciate all of the comments, which have improved the paper. Our point-by-point responses are detailed below.

RC – Review Comments; AC – Authors Comments

RC: (1) The paper discusses a lot the mathematical effects of the parameterisations, but does not go into detail which physical concepts are accurately captured by the individual approaches, e.g., it is not obvious why the formulas for E result in differences of more than one order of magnitude. The only aspect which is elucidated are differences

resulting from particle types and corresponding terminal velocities, but they cannot explain the differences between the three formulations for E, but only the differences among the individual E values for each respective scheme.

Also it is not obvious, why the differences are larger for the lower snow intensities than for stronger solid precipitation.

This is similarly true for the individual parameterisations for Vd and A; the effects are well described and the subsequent influence on the scavenging efficiency is obvious, but the physical (not the mathematical) reasons of the different formulations for the parameters remains unclear.

AC: We agree with the reviewer's concern. Consequently, we have added a few sentences to explain the physical concepts captured by each semi-empirical formula for E and how they are parameterized. We realize that there are some differences between each formula, and sometimes these differences are large. Since the main objective of the paper is to review and quantify the uncertainties that arise from different treatments of each of the key parameters associated with the calculation of the size-resolved scavenging coefficient, we included three commonly used formulas in the paper.

The following discussion was added at the end of Section 2.1: "Generally these formulas use a conceptual model that a hydrometeor can collide with an aerosol particle through the mechanisms of Brownian diffusion, interception, and impaction. Both the formulas of Slinn (1984) and Murakami et al. (1985) consist of three terms, representing the contributions from these three mechanisms respectively. Dick's formula has only two terms, considering the contributions from Brownian diffusion and impaction but neglecting interception. All three formulas parameterize the contribution from impaction using the Stokes number. The contribution from collisions due to Brownian diffusion is parameterized using the Schmidt number by Slinn, the Schmidt number and the Reynolds number by Murakami et al., and the Reynolds number and the Peclet number by Dick. Slinn's formula parameterizes the contribution due to interception through the Reynolds number and the interception parameter, while Murakami et al. only use the interception parameter and parameterize it using a simple power-law relationship."

The differences in E values between the different E formulas are larger for smaller collectors than for larger collectors (Fig .1). For any snow particle size distribution shown in Fig. 3, lower snow intensities would have more small collectors than stronger snow intensities. Thus, the differences in the scavenging coefficient that arise from using different E formulas are larger for lower snow intensities (compare the ranges of solid liens and dashed lines in Fig. 2). A brief explanation was added in Section 3.1 in the revised paper.

Most VD and A formulas are empirical (Fig. 5), and both quantities are parameterized as simple functions of snow particle diameter (in water equivalent). The physical reasons for the differences in scavenging coefficient are simple. The faster the falling speed and the larger the cross-sectional area of a collector, the faster the collection process will happen. Thus, formulas giving larger VD and A will result in larger scavenging coefficients. A brief explanation was added in Section 3.3 in the revised paper.

RC: (2) In the paper the terminology for snow / ice crystals and solid hydrometeors in precipitation is not always clear. To certain degree the different snow crystal types are explicitly considered in the formulations, but in Fig. 10 it is not clear if the shape is still considered.

However, typical regional to global scale models do not provide the information about the crystal type and shape, but only total solid precipitation flux, which then can be used for the scavenging calculations either with additional assumptions on crystal type distributions or by using generalised crystal types/shapes. This should be discussed in some detail if the applicability of the parameterisations is suggested.

AC: The term "snow crystals" is now used consistently in the revised paper. Figure 10 shows the combined range of scavenging coefficient presented in the four panels of

Fig. 8 and thus covers all four of the different snow particle shapes. This has been made clear in the revised caption of Fig. 10.

We have also added a brief discussion in Section 2 in the revised paper on the availability of precipitation information in regional- to global-scale models and potential uncertainties caused by different assumptions about snow particle shape.

RC: (3) As the empirically fitted formula of Paramonov et al. includes all processes like the electric charges, thermophoresis, etc. it is reasonable that the obtained values for are larger than in the conceptual approaches, in which those processes are neglected. Even though they are assumed to have small influences only, close to the minimum values they will potentially have the largest importance. This becomes most obvious in Fig. 7, where theoretical approaches underestimate the observed fit.

Furthermore, the turbulence during the snow events can likely cause a completely different spectrum of terminal velocity especially for dendrite snow flakes, such that the effective scavenging can be much larger than theoretically assumed. This should be considered in the comparisons.

However, it should be taken into account, that the observations are an empirical fit to a multitude of individual events and do not represent the prescribed settings as for the theoretical approaches.

AC: We completely agree with this comment. In fact, a number of previous studies, including a study by ourselves (e.g., Andronache et al., 2006; Wang et al., 2011; Quérel et al., 2013), demonstrated that many processes could have contributed to the overall scavenging observed under field conditions but were not included in the theoretical framework for the scavenging coefficient. Some of these processes, however, may not be needed in theoretical formulations of scavenging coefficient because they are included elsewhere in large-scale models, but some should be considered (such as those factors increasing the collection efficiency). We have added a new paragraph in Section 4.2 to address this issue.

RC: (4) Would the diversity even increase if also the terminal velocity is calculated with a different scheme than the Mitchell and Heymsfield approach? This is not discussed in the manuscript, but as this quantity alone can influence the values for one E by up to one order of magnitude (Fig. 7), I am surprised that the overall diversity remains smaller than 2.5 orders of magnitude, especially if the combined uncertainty is stated to be larger than the sum of individual uncertainties (Page 14840, line 22).

On the other hand, to which degree do these effects cancel out, since for increased snow rate the differences from E are reduced, but from N(dp) are growing?

AC: In a separate study following the present study, we aimed to develop a simple new parameterization for below-cloud scavenging by snow and rain. In the new study, a full set of sensitivity tests was conducted using all available combinations of the formulas listed in Tables 1-4 over a range of precipitation intensities from 0.001 mm h-1 to 10 mm h-1. Based on this new set of sensitivity tests, we did find that the uncertainties shown in Figs. 7 and 8 increased slightly in most cases. More specifically, for a spherical snow particle shape (Figs. 7a and 8a), nothing changed since only one formula is available for this shape (see Table 3). For dendrites (Figs. 7b and 8b), the uncertainties increased by less than a factor of 2 under heavy snowfall conditions (e.g., 10 mm h-1), which also agrees with the findings shown in Fig. 6, where scavenging coefficient generated by using empirical VD formulas is slight lower than that from using theoretical VD formula. The same conclusion was found for columnar shapes (Figs. 7c and 8c), although the minimum of scavenging coefficient shifted down a bit more than for the dendritic shape (see Fig. 6). For graupel (Figs. 7d and 8d), the maximum scavenging coefficient shifted up by less than a factor of 2 (which agrees with those curves shown in Fig. 6). We have added a short discussion at the end of Section 4.1 based on above results.

In general, as we discussed in Section 3.3, VD alone can create uncertainties in scavenging coefficient of only a factor of 2 to 3 for the same snow particle shape for all aerosol particle sizes. However, if different snow particle shapes are considered, then

the combined uncertainties will include a contribution from A and can thus be as high as a factor of 10. Since we did not include the VD impact on scavenging coefficient (all profiles are calculated using the same theoretical VD formula of Mitchell and Heymsfield) in Fig. 8, the overall diversity remains smaller than 2.5 orders of magnitude.

The degree of cancellation varies with different cases (snow particle shape, precipitation intensity, aerosol size, etc.). If the two lines from different combinations of formulas were very close, then everything cancelled out. But if the two lines were further apart, then the different factors enhanced each other in the overall uncertainty.

RC: (5) How is the integral over the collector sizes in the respective size distribution discretised for the numerical calculations? This should be mentioned in the manuscript, as the impact of the collector size distribution will be larger for a highly resolved discretisation in solid hydrometeor size, but on the other hand, this will make the calculations computationally more expensive and less suitable for large-scale, long-term simulations.

Similarly, it is not described how the aerosol spectrum in Sect. 4.3 is discretised to calculate the loss in mass and number concentrations due to the scavenging. Are the 100 size bins (Page 14842, line 19) assumed for the aerosol or the precipitation distribution (or both or are they overlapping)?

AC: In large-scale models, a small number of size bins (e.g., <20) is usually used due to computer time constraints. However, this was not an issue in the present study so we used 100 size bins to discretize both the aerosol-particle and snow-particle size distributions. We have added information on size bin and structure and on the initial aerosol size distributions used for producing Fig. 9 in Section 4.3 of the revised paper.

Minor comments: RC: (1) Please correct the unit in the caption of Fig.2: "m" should be "mm"

AC: Corrected.

RC: (2) Abstract last sentence: This does not become obvious from the manuscript in its current form. Consequently, this sentence should be reformulated less strongly.

AC: The statement referred to is based on the discussion presented in Section 4.4. As mentioned above, in a separate study following the present study, we conducted more sensitivity tests and compared scavenging coefficients under snow and rain conditions. Based on the median and the upper-range values of scavenging coefficients produced from all available theoretical formulas, it is very likely that scavenging of aerosol by snow is faster than by rain. In our opinion, the current statement is not a firm conclusion, but a speculation supported with some limited evidence (cf. Sec. 4.4), and an appropriate caution to that effect is included in the existing statement.

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