Responses To Referee #3

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

RC- Review Comments; **AC** – Authors Comments

RC: General Comments:

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).

AC: We agree with the comment that theoretical formulations for scavenging coefficient should not be expected to agree exactly with field measurements. In fact, a number of earlier studies, including a study by ourselves (e.g., Andronache et al., 2006; Wang et al., 2011; Quérel et al., 2013), demonstrated that many factors could have contributed to the overall scavenging observed under field conditions but not all of these factors were included in the theoretical scavenging-coefficient formulations. Some of these processes, however, may not be needed in the theoretical formulations for scavenging coefficient because they are accounted for elsewhere in large-scale models (Wang et al., 2011), but some should be considered (such as those factors increasing the collection efficiency) (Quérel et al., 2013). We have added a new paragraph in Section 4.2 to address this issue directly. As well, assumptions for the empirical parameterization were modified to be more close to real conditions, following point 4 and some discussions were also revised based on other detailed comments provided below.

Specific Comments:

RC: (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.

AC: We agree with this comment. We have chosen to use the term “snow crystals” throughout the discussion in the revised manuscript. Information about the size range and size-bin number considered have also been added in the revised paper. Briefly, 100 size bins were used to discretize the size distribution of snow particles and a second set of 100 size bins were used to discretize the size distribution of aerosol particles. The size ranges considered were 10 μm to 10 mm in water-equivalent particle diameter for snow particles and 0.001 μm to 100 μm in particle diameter for aerosol particles. A constant volume ratio between successive size bins was used for both discretizations.

RC: (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 dp swept per unit time across an area. . ..’?

AC: Agreed. This is a helpful clarification.

RC: (3) Page 14833, line 14-15: Perhaps consider adding a reference related to the importance of electric charges in the 0.1-1 μm size range e.g. Tingsley et al., 2000.

AC: The reference has been added as suggested.

RC: (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 Quérel 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.

AC: We suspect that the dependence of snow scavenging of atmospheric aerosols on ambient RH could be because the higher the RH, the stickier the snow particles will be. We have added this speculation in the revised paper and have also pointed out the differences between the sets of mechanisms considered in the theoretical formulations and the field experiments. As mentioned above, a paragraph has also been added in Section 4.2 to discuss in some detail the potential causes of the discrepancies between theoretical results and field measurements.

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

AC: Yes, we have made a full set of sensitivity tests using all possible combinations of the available formulas listed in Tables 1-4. We chose to show only one set of V_D and E and to focus on discussing the uncertainties caused by various $N(D_p)$ formulas in Fig. 4. The magnitudes of the uncertainties caused by various $N(D_p)$ formulas are similar if other formulas are used for V_D and E, as can be seen from Figures 7 and 8. For example, the black lines in Figs. 7 and 8 show the sensitivity of Λ_{snow} to the different $N(D_p)$ using the formulas of Slinn (1984) for E and the formulas of Mitchell and Heymsfield (2005) for V_D while the blue lines show the results of using the formulas of Dick(1990) for E and the formulas of Mitchell and Heymsfield (2005) for V_D. The V_D formula of Mitchell and Heymsfield (2005) was chosen for Fig. 4 because it is the only formula that is applicable to all snow particle shapes according to the original literature (as is pointed out in Section 4.1).

RC: (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?

AC: It should not be a linear increase because different $N(D_p)$ formulas respond differently to increasing snow intensity. For example, the total snow particle number concentrations from the MP and SC distributions increase and those from the SS distribution decrease with increasing snowfall intensity due to different N_{0e} used in these formulas (Table 5). The differences in the total number concentrations from different size distributions can change from less than one order of magnitude to more than two orders of magnitude when snowfall intensity increases from 0.1 to 10 mm h^{-1}. In addition, although all $N(D_p)$ profiles shift to larger snow particle sizes when snowfall intensity increases, the rates of increase of the largest snow particles (> 1 mm) are different (Table 5).

As mentioned above, we conducted a full set of sensitivity tests covering all possible combinations of available formulas under 37 precipitation intensities from 0.001 mmh^{-1} to 10 mmh^{-1}. Examination of the sensitivity test results confirmed the above suggestion.

RC: (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?

AC: We think the reviewer may have intended to refer to page 14837 instead of page 14839. Again, we took one size spectrum as an example. Similar conclusions can be generated from using other size spectra.

RC: 8) Page 14840, lines 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 size range or related to a specific set of parameterizations.

AC: The statement is intended for all aerosol size ranges, although this phenomenon is not so apparent for the largest aerosol sizes. Uncertainties shown in Figs. 7 and 8 represent the range of different combinations of the available formulas and thus are not associated with any specific set of parameterizations.

RC: (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.

AC: This suggestion requires comparing many different lines and will end up providing little useful information, in our opinion.

RC: (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}?

AC: The following figure shows the corresponding results for snowfall intensities of 10.0 mm h^{-1}. In comparison with the results in Fig. 7 (for snowfall intensities of 0.1 mm h^{-1}) and Fig. 8 (for 1 mm h^{-1}), all Λ_{snow} values increase further for all aerosol particle sizes with the increase of snowfall intensity and the magnitude of uncertainties in Λ_{snow} values increases as well.

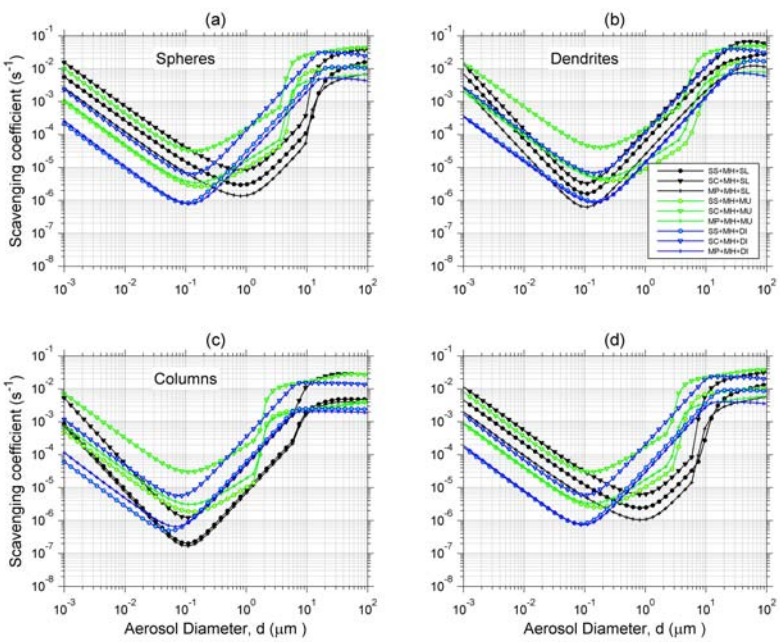

Figure. Same as in Fig.7 but for a snowfall intensity of 10.0 mmh^{-1}

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

AC: We compared the ratio of the maximum to the minimum Λ_{snow} for all aerosol sizes using data in both Fig. 7 and Fig 8. We found that the ratio increased by 30%-60% for ultrafine aerosol particles ($d_p < 0.01$ μm), 10%-25% for particles in the size range from 0.01 μm to 10 μm, and 15% for very large particles ($d_p > 10$ μm) when the snowfall intensity increased from 0.1 mm h^{-1} to 1.0 mm h^{-1},. We have added this information in Section 4.1 in the revised paper

RC: (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.

AC: The RH value was set as 90% in the calculations, consistent with the discriminant RH value mentioned in the original paper.

RC: (13) Page 14841, line 18-20: If I understand correctly, the Kyrö 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.

AC: Agreed. Results from the Kyrö et al. (2009) parameterization have been added in the revised paper for comparison purposes, and related discussion has also been added in Sect. 4.2.

RC: (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.

AC: Thanks very much for pointing this out. We have modified the assumption about the snow intensity frequency distribution to be closer to the actual field conditions. Because weaker snow intensities (0.1-0.2 mm h^{-1}) occurred more frequently than stronger snow intensities, this better supports our discussion presented in the paper (i.e., the values from the empirical formula were more representative of the cases shown in Fig. 7 and should be shifted up significantly in Fig. 8).

We suspect that one of the major reasons that Paramonov et al. (2011) found a low correlation between snow intensity and the scavenging coefficients was the small range of snow intensities sampled during the measurement period.

RC: (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 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.

AC: As we responded to the general comment, we were aware of the set of mechanisms considered in the theoretical formulas vs. the greater number that influence field measurements. Although we do not expect complete agreement between theoretical and field-generated values, the field measurements serve as constraints (i.e., upper bounds) to the theoretical formulas. If we know what mechanisms would increase the collection efficiency but are not included in the theoretical formulas, we would expect a certain degree of discrepancy. For example, vertical diffusion and the subsequent in-cloud scavenging would increase collection of aerosol particles observed in the field (Andronache et al., 2006; Wang et al., 2011); theoretical values would thus be expected to be smaller than field measurements (as is the case shown in the comparison in the present study, see Fig. 7). These comparison results would suggest that the existing formulas might be appropriate for large-scale model applications because those processes causing the discrepancy are included in the continuity equation in large-scale models (Wang et al., 2011). On the other hand, some mechanisms might increase collection efficiency but are not considered in either theoretical formulations of scavenging coefficient or in large-scale models. For example, Andrea Flossmann's group observed the increased collection efficiency due to the rear vortex that develops behind a falling raindrop (Quéerel et al., 2013). In this case, modification/improvement of existing theoretical formulas might be needed.

We have added some discussion based on above information and the reviewer's comment in the revised paper.

RC: (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.

AC: More information has been added in the revised paper, including the initial size distribution of the aerosols. We are reluctant to provide too much detailed information here because this figure used the same approach as that used in Wang et al. (2010) and we referred the reader to that paper for more details.

RC: (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.

AC: Small aerosols dominate number concentration and large aerosols dominate mass concentration. Large aerosols have higher scavenging coefficients than smaller aerosols. This explains why mass concentration decreased more rapidly in the first hour than the number concentration, but the trend reversed in later hours. Thus, the impact of using different scavenging coefficient formulas was different for number and mass concentrations. A brief explanation has been added in the revised paper. Again, we prefer not to provide too much detailed discussion here because a detailed discussion was presented in Wang et al. (2010) for rain cases, and we referred the reader to that paper for more details.

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

AC: We meant both, and this has been made clear in the revised paper.

RC: (19) Page 14844, lines 22-24: As indicated in point 14), I am not sure that this adjustment to the scavenging coefficients is well justified.

AC: See our response to point 14 above.

RC: (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?

AC: The new parameterization should not be based on field measurements (as pointed out in the response to point 14 above). The new parameterization is still based on existing theoretical formulas. Instead of assuming all component parameters (E, V_D, A, $N(D_p)$), the new formula is a semi-empirical fit of theoretically-generated scavenging coefficient values and thus is much easier to use in large-scale models, although we do admit that the new semi-empirical formula has similar uncertainties to existing theoretical formulas.

RC: (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?

AC: The ranges and trends of uncertainties are similar if Slinn's formula for E is used, although the shape of the curves changes (see the figure below).

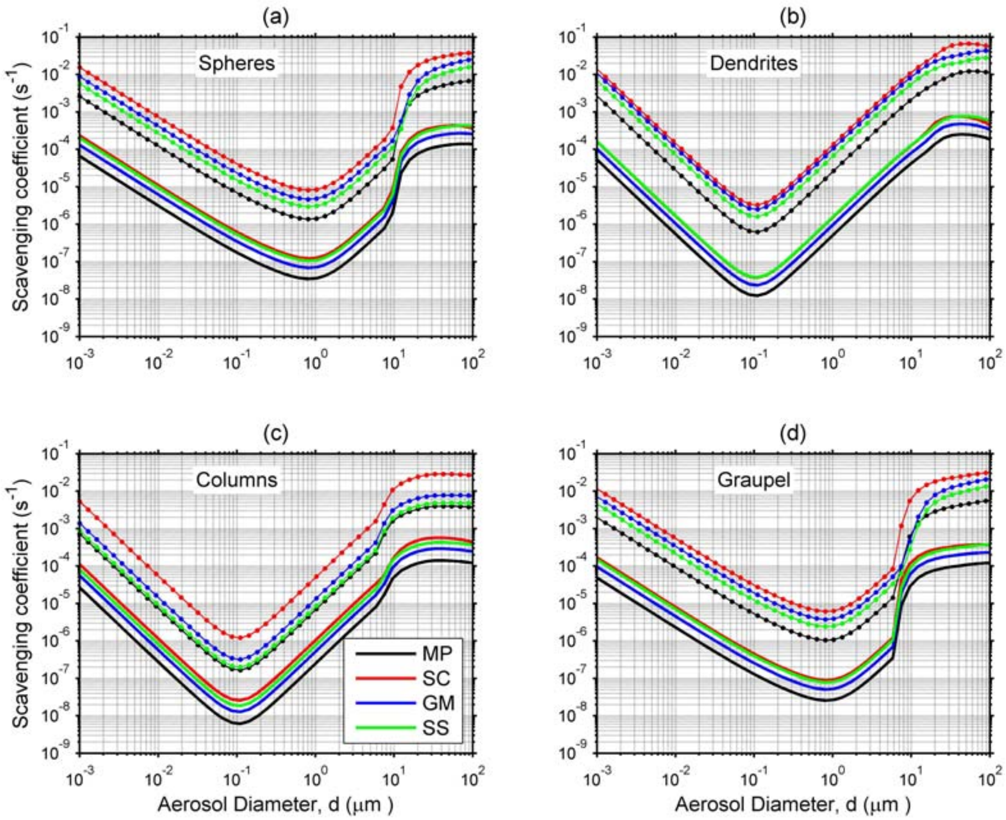

Figure. Same as in Fig.4, but using Slinn (1984) $E(d_p, D_p)$ parameterization (note the change in y-axis range)

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

AC: See our response to comment 10 above.

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

AC: We have added the phrase "based on the median and upper range of theoretically generated values." We think the upper range of theoretical values is closer to reality because this range is closer to, though smaller than (as expected), field-generated values. More information is provided in our response to the next comment.

RC: (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.

AC: We do not expect that they will agree with each other. Instead, we expected that the theoretical values should be somewhat smaller than field values due to the various factors mentioned above (see our response to point 15 above). The comparison results presented in this study meet our expectations and the differences between theoretical and field values can be mostly explained by known factors. This point has now been made clear in the Abstract and in the text in the revised paper.

RC: (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.

AC: We used the upper and lower ranges of scavenging coefficients shown in Figs. 7 and 8 to produce Fig. 9. Thus, the range of scavenging coefficient differences can be identified from those figures, and they differ by around two orders of magnitude differences. This is now pointed out in the revised paper.

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

AC: We have added one sentence to the Abstract related to this comment:
"Assumption of different snow particle shapes (and thus different Vd and A) will cause an uncertainty of up to one order of magnitude in the calculated scavenging coefficient."

RC: (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.

AC: While there are other approaches to represent below-cloud scavenging (see a review in Gong et al., 2011 for approaches used in regional- to global-scale models and theoretical studies by Flossman's group), we believe the common formulas used for calculating snow scavenging coefficients have been included in the present study.

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

AC: We are aware that both of the terms "below-cloud scavenging" and "impaction scavenging" have been used in the literature, but we believe that the former is more common than the latter. Considering that in the E formulas the collection mechanisms also use the word "impaction", we have chosen to add "below-cloud" to the title to avoid any confusion.

RC: Technical Corrections:

AC: We have revised the figures and figure captions as suggested and we have added the recommended references.