Dear Editor,

We have carefully addressed both reviewers’ comments and have revised the paper accordingly. Our detailed point-by-point responses are attached. For your and the reviewers’ convenience to review our revisions, we highlighted major changes in the revised apper. We hope you and the reviewers will find the revised paper meets the standard of the journal.

Thank you very much for editing the paper.

Sincerely,

Leiming Zhang and coauthors

Reply to Reviewer #1

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

RC - Review Comments; **AR** – Authors's Responses

General Comments:

RC: My major concerns with the manuscript in its present form are that 1) it does not give a reader a clear indication of the conditions when the new parameterization might be most suitable and 2) the use of the 90th percentile of coefficient values is not well justified, nor are alternatives examined. While theoretical and measurement based scavenging coefficients are expected to differ, it is not well established that theoretical values in the 90th percentile should give a more accurate representation of reality simply because they are closer in magnitude to the measurement based values, as outlined below.

AR: The scheme developed in this study is intended for use in a wide range of chemical transport models (CTMs), which often have limited characterizations of precipitation processes and may not be able to distinguish between different rain types or droplet size distributions. The scheme only requires information about precipitation intensity and precipitation type, two basic properties of precipitation that any CTM should have information about. Thus, there is no practical need to specify precipitation conditions for which the scheme is most suitable. Besides, the scheme was developed from a combination of all scavenging existing schemes and thus should not only be applicable to certain specific precipitation conditions.

The choice of the 90^{th} percentile of the ensemble of the existing theoretical-formulas-generated scavenging coefficient values was based on conclusions and recommendations presented in our three previous studies (Wang et al., 2010, 2011; Zhang et al., 2013). This choice was believed to give smaller uncertainties than other alternatives. However, this issue is addressed further in the responses provided below, particularly in responses 1, 7, and 15.

Specific Comments:

RC: (1) The third paragraph of the introduction states that the upper range of available scavenging coefficient theoretical formulations are thought to be more realistic because they are closer to, while still smaller than the field derived estimates. While I concur that the theoretical formulations and field measurements for the scavenging coefficient are expected to differ, the magnitude of this difference in the case that the theoretical formulation was perfect is not well established since the measurement based values do include processes (including storm dynamics) not expected to be included in the theoretical formulations. Thus I am not convinced that the development of a parameterization based on the 90th percentile of theoretical formulations is fully justified. Are the authors able to provide any additional support for this approach, particularly for snow, or at least give a more thorough consideration of alternative assumptions?

AR: As mentioned above, a detailed comparison between existing theoretical formulas and field data was presented in Wang et al. (2010) for rain-scavenging cases and in Zhang et al. (2013) for snow-scavenging cases. Sensitivity tests were also conducted on the impact of one process (vertical diffusion) that influenced the field data but which was not included in the theoretical formulas (Wang et al., 2011). We do agree that there are many other factors, such as storm dynamics (e.g., Chate, 2005, *Atmos. Environ.*, 39, 6608-6619), as mentioned by this reviewer, and rear capture of particles by falling drops (Quérel et al., 2013, *Atmos. Res.*, in press), that are not explicitly considered here. These factors need more investigation and may not be easily included in a simple scheme such as the one proposed here. We have added a short comment to the revised paper to note the neglect of these other factors (in Sect. 1).

For the cases of rain scavenging, Fig. 1d of this study shows that 50th-percentile values of the ensemble of theoretical Λ_{rain} curves are nearly two orders of magnitude lower than the values obtained from the majority of the field data, whereas the 90th-percentile values are around one order of magnitude lower than the field data. Is there a reason to choose between these two curves? On the one hand, consider that the scheme has been developed for application in CTMs and that some processes that influenced the field data have also been included separately in the CTMs, suggesting that Λ_{rain} values from the new scheme should be somewhat smaller than the field-data generated values, while on the other hand, a number of factors that can potentially enhance particle collection (such as those mentioned above) are not considered in any of the theoretical formulas, so that the existing formulas are likely to be biased low. But in addition, the results of the sensitivity tests conducted by Wang et al. (2011) showed that accounting for turbulent diffusion could increase Λ_{rain} values by one to two orders of magnitude, enough to bring the *upper end* of the theoretical Λ_{rain} values into agreement with field measurements. Thus, considering that the 90th-percentile values are still one order of magnitude lower than the field data and that the various theoretical formulations cannot all be correct, we believe that the 90th percentile of the theoretical Λ_{rain} curves should be more representative than other lower values, e.g., 50-80th percentiles.

For the cases of snow scavenging, the differences between existing theoretical formulas and field data are not as large as for the case of rain scavenging, but the 90th-percentile theoretical values are still smaller than the field-data-derived values. To be consistent with the rain case, the 90th percentile has also been used for the snow case. We agree that scavenging coefficient values from the 50-90th percentile range may be as representative as the 90th percentile for snow scavenging, but there is no way at this time to validate this assumption due to the very limited field data that are available.

RC: (2) Does the 90th percentile of theoretical formulations for both rain and snow correspond primarily to a certain combination of the existing theoretical formulations? This was not clear in the text but would be helpful to know the combination used.

AR: No, the 90th-percentile Λ profiles for rain and snow do not exactly match any of the theoretical formulations shown in the paper, but they are close to certain combinations. For rain scavenging, the 90th-percentile Λ_{rain} values are close to either the combination of using the $E(d_p, D_p)$ formula of Andronache et al. (2006) (Table 1) with any of the gamma or lognormal distributions for $N(D_p)$ (Table 2) or the combination of the $E(d_p, D_p)$ formula of Slinn (1984) (Table 1) with any of the exponential distributions for $N(D_p)$ (Table 2). The choice of terminal-velocity formula (Table 3) made little difference. For snow scavenging, the 90th-percentile Λ_{snow} values are close to the combination of the $E(d_p, D_p)$ formula of Murakami et al. (1985 (Table 4) with the $N(D_p)$ formula of Scott (1982) (Table 5) for any type of snow particle (Table 6). We have added a short note on this finding in the revised paper (see Sect. 3.1).

RC: (3) The last paragraph of the methodology states that steps 1 and 2 of the parameterization development were only conducted for precipitation at 1 mm hr-1. If other precipitation rates were considered, would this change the values retained as 'realistic'?

AR: During the development of the new Λ parameterization, we have tested steps 1 and 2 using different precipitation intensities. We found that the patterns of the Λ profiles at different precipitations are very similar to those in Fig. 1 (see also Fig. 8 of Wang et al., 2010). Thus, the choice of various combinations of product terms for another precipitation intensity would be the same as using the precipitation intensity of 1 mm h^{-1}. We have clarified this point in the revised paper (see Sect. 3.1).

RC: (4) The first paragraph of Section 3 states that a fixed ambient temperature and pressure was assumed. How might this influence the performance of the parameterization under different conditions?

AR: We have conducted sensitivity tests using different temperature and pressure values to address this comment. Temperature values of 5°C and 30°C for the rain cases and -5°C and -30°C for the snow cases and an ambient pressure value of 900 hPa for both the rain and snow cases were used in the sensitivity tests. The differences caused by using different ambient temperature and pressure values are generally within 10% of the previously reported results for all particle sizes for both rain and snow scavenging; the only exception is for rain scavenging of particles of sizes 0.1-2.0 μm, for which the bias can be up to 30%. We have presented these results in a new figure, Figure 8, and we have added a summary of the results of these sensitivity tests and related discussions in the revised paper to a new section, Section 4.3.

RC: (5) The definition of the raindrop size (page 5907, line 19) seems unusual. Hydrometeors of 1 um in diameter would usually not be considered raindrops.

AR: We agree with the reviewer that a hydrometeor of diameter of 1 μm would normally be described as a very small cloud droplet or ice particle. The definition used in the manuscript was only for the convenience of numerical calculation. In fact, droplet concentrations at small droplet sizes are actually negligible and have little impact on the final results. We performed a sensitivity test by reducing the hydrometeor diameter range from [1 μm, 10 mm] to [10 μm, 10 mm] while keeping the same number of size bins and found no noticeable impact on the simulation results.

RC: (6) The first paragraph of Section 3.1 states that the component parameter formulas come from a wide range of rain types. Does the choice of the 90th percentile yield a selection of one rain type primarily? In that case, what rain type was primarily used on the parameterization development? This information could be helpful to the reader in understanding the parameterization.

AR: As explained in our responses to comment 2 above, this choice does not match any specific combination of component formulas nor any specific rain type. As also explained in our responses to the general comments, the new scheme is aimed to be applied in a wide range of CTMs, few of which have a specification of rain type.

RC: (7) The agreement of the 50th to 90th percentile of theoretical formulations with the experiment of Sparmacher et al. (1993) seems to suggest that a parameterization based on the 50th percentile might be equally reasonable. Do the 50th percentile values correspond to any certain rain type or combination of the input formulations that might lead to a preferential choice of the 50th percentile under certain conditions for alternative parameterization development?

AR: No, the 50^{th}-percentile Λ profiles for rain and snow do not exactly match any of the theoretical formulations but they are close to certain combinations. For rain scavenging, the 50^{th}-percentile Λ_{rain} values are close to the combination of the $E(d_p, D_p)$ formulas from either Slinn (1984) or the updated Ackerman et al. (1995) formula (Table 1) with gamma or lognormal distributions for $N(D_p)$ (Table 2). For snow scavenging, the 50^{th}-percentile Λ_{snow} values are close to the combination of (a) the $E(d_p,D_p)$ formula of Slinn (1984) (Table 4) with the $N(D_p)$ formula of Sekhon and Srivastava (1970) (Table 5) for dendrites and columns (Table 6), (b) the $E(d_p, D_p)$ formula of Murakami et al. (1985) (Table 4) with the $N(D_p)$ formula of Sekhon and Srivastava (1970) (Table 5) for columns and spheres (Table 6), and (c) the $E(d_p, D_p)$ formula of Dick et al. (1990) (Table 4) with the $N(D_p)$ formula of Sekhon and Srivastava (1970) (Table 5) for dendrites (Table 6).

It is also important to remember that the controlled experiment was likely to results in underestimates since some factors that should have played a role in the collection process were excluded in the experiment. We should favour agreement with the majority of the field experiments. Also see our responses to comment 1 above.

RC: (8) The second paragraph of page 5912 discusses the abrupt change in the A(d) and B(d) values at particle sizes between 1 and 2 um. Are you able to provide any insight on the physical basis for the abrupt shift?

AR: The scavenging process is governed by interactions between aerosol particles and hydrometeors, which are influenced by Brownian diffusion, interception, inertial impaction, thermophoresis, diffusiophoresis, airflow turbulence, electrostatic attraction, etc. Brownian diffusion dominates the collection of smaller particles, in particular ultrafine particles (d < 0.01 μm), but the role of this mechanism decreases rapidly with increasing particle size. Inertial impaction, on the other hand, becomes significant when the particle diameter is larger than a few microns, e.g., 3 μm for unit-density particles and a 1 mm raindrop (Phillips and Kaye,1999; Loosmore and Cederwall, 2004), where the particle Stokes number (*St*) is greater than the critical Stokes number (St^*). For particles in the diameter range from 0.01 to 3 μm, on the other hand, although more mechanisms (e.g., interception, diffusiophoresis, thermophoresis and electric charges) play a role in the collection process, the overall contribution of all of these mechanisms result in scavenging coefficients that are two orders of magnitude lower compared to contributions of the Brownian diffusion and nertial impaction to ultrafine and large-particle scavenging, respectively. As a result, the combined action of the microphysical processes lead to very efficient scavenging of ultrafine particles and large particles but to weaker scavenging for particles with diameters in the range from 0.01 μm to a few micrometers, which is commonly called the "Greenfield gap" or "scavenging gap" range.

From Eq. (5), we can see that the scaling factor *A(d)* corresponds to the scavenging coefficient at particle diameter *d* when precipitation intensity R = 1.0 mmh^{-1} and the exponent term *B(d)* represents the rate of change of Λ to changes in R for each particle diameter *d*. The abrupt change in the values of *A(d)* and *B(d)* at particle diameters between 1 and 2 μm is associated with the transition of the dominant collection mechanism from the multiple mechanisms within the "Greenfield Gap" to the inertial impaction mechanism for large particle sizes.

RC: (9) The last sentence of Section 3.1 states that the uncertainties associated with the new scheme should not be larger than the uncertainties with the existing parameterizations (which are in the order of magnitude range). This is fair enough – the new scheme does not accomplish any reduction in uncertainty in the scavenging coefficients but does provide a convenient parameterization as a function of quantities that are predicted by a global model. What needs to be clear to the user of such a parameterization is whether the parameterization favors a certain rain type or set of physical conditions. Could this information be presented more explicitly?

AR: As explained above, the new parameterization is aimed for general application, not for specific rain types. Moreover, it does not exactly match any of the Λ profiles derived from certain combinations of the existing theoretical formulations and it does not appear to favour any particular rain type or set of physical conditions (see response 2). It is worth noting as well that as a consequence of the development methodology, this parameterization does represent the upper range of an ensemble of the existing theoretical $\Lambda(d)$ values for all particle diameter values, which provides some additional information about the uncertainty associated with the scheme. Some text has been added at the end of Sect. 3.1 to make these points.

RC: (10) Section 3.2 deals with the parameterization for the snow scavenging. My previous point for rain applies also to the presentation of the snow scavenging parameterization. Is the parameterization (developed by choosing the 90th percentile values) most applicable for certain types of snow and habits?

AR: As explained above (see response 7), the new scheme does not exactly match any of the Λ_{snow} profiles derived from certain combinations of the existing theoretical formulations, and it was not developed for specific snow types. It can be implemented in a CTM without needing any information about snow type (which is often not available).

RC: (11) Section 3.2 states that no unrealistic values were found and excluded from the ensemble of scavenging coefficients. Figure 4a does appear to show clusters of the lines around two distinct minima (similar to Fig. 1a). Can the authors give explanation why all values were considered realistic for Fig. 4a but not for Fig. 1a?

AR: These two clusters with two distinct minima were caused by applying different formulas to different snow particle shapes, and should not be considered as unrealistic (cf. Figs. 1, 2, and 8 of Zhang et al., 2013). Given that information about snow particle shape is often not available in CTMs, however, we deliberately grouped all of the existing formulas together without explicit consideration of snow particle shape. A note on this decision has been added to Sect. 3.2 of the revised manuscript.

RC: (12) I found the justification of the use of the 90th percentile to be weaker for snow than for rain since there is even less experimental data available (Fig. 4b compared to Fig. 1d) – thus additional information to characterize the conditions most applicable for the snow parameterization (point 10 above) would be particularly helpful.

AR: We agree this comment, and we noted this limited availability of experimental data in the last paragraph of the Conclusions section. More experimental studies for a wider range of locations and meteorological conditions are indeed needed to validate this choice. But as mentioned above, the parameterization has been developed for general applications in CTMs and not for any specific snow types. We have added a note regarding this comment in Sect. 3.2 of the revised manuscript.

RC: (13) Also, how does the exclusion of unrealistic values based only on one precipitation intensity for snow influence the results? Have you checked other precipitation intensities?

AR: Our response to comment 3 above also applies to the snow case.

RC: (14) Section 4.1 gives a comparison with previous B(d) values – perhaps a table might helpful to see these values more readily. Some of these previous values are related to field measurements and others are related to theoretical formulations – would you expect to be able to compare equally between the two? Also, are you able to make any comparisons related to A(d)?

AR: Thank you for this comment. We have added a table (Table 9) and related discussion in the revised paper to address both B(d) and A(d) (see Sect. 4.3).

RC: (15) Related to the comparisons in Section 4, if you formulated the parameterization based on the 50th percentile or any other percentile as opposed to the 90th percentile, would the agreement of B(d) with previous studies be any better or any worse?

AR: To address this comment, we repeated all of our development steps using the 50th percentile in place of the 90th percentile. The results are shown in the following Fig. A for rain scavenging and Fig. B for snow scavenging. We can see that the value of *B(d)* based on 50th percentile parameterization are in the range of 0.70-0.88 for rain scavenging (Fig. Ad) and 0.64-0.90 (Fig. Ad) for snow scavenging. There is no dramatic change in the *B(d)* values compared to those using the 90th percentile data, and thus, the comparison of *B(d)* with literature values would show similar results.

RC: (16) In Section 4.2, the ratio between the snow and rain coefficients is noted to change with particle size. Are you able to add any comments on the physical mechanisms that drive these changes?

AR: Figures 1d and 4b show that the 90th-percentile Λ profiles are qualitatively similar for the rain and snow cases. However, two significant differences exist between these two profiles. The first difference relates to the value of the aerosol particle diameter at which the minimum Λ value occurs. The Λ_{rain} minimum occurs at a particle diameter around 0.4 µm whereas the Λ_{snow} minimum occurs at a particle diameter around 0.1 µm (which corresponds to a local minimum in Fig. 7). For submicron particles, scavenging is mainly controlled by the interception mechanism and the contribution to scavenging due to interception increases with increasing particle diameter. For snow scavenging, the increase of scavenging coefficient with particle diameter in this size range is faster than that for rain scavenging due to the larger cross-sectional areas of snow particles. Thus, the ratio between the snow and rain scavenging coefficients in Fig. 7 increases in the particle diameter range between 0.1 µm to 1.0 µm. The second significant difference relates to the abrupt transition of Λ_{rain} from an interception regime to an inertial-impaction regime at a particle diameter of about 2 um. For particle diameters larger than 2 um, Λ_{rain} increases more quickly with *d* than Λ_{snow} does. As a result, the Λ_{snow}: Λ_{rain} ratio decreases quickly with increasing *d* until leveling off for particle diameters close to 10 um. A paragraph discussing for this particle-size-dependent behaviour has been added in the revised paper (see Sect. 4.2).

RC: (17) The conclusion starts by mentioning that the use of existing theoretical formulations for the scavenging coefficients requires somewhat arbitrary choices to be made. However as you note at page 5906 line 27, the choice of the 90th percentile is also somewhat arbitrary. This led me to think that in order for this new parameterization to be a step forward in any manner other than simply being a more convenient formulation – the text should give as much understanding as possible about the range of conditions when these rain and snow parameterizations are considered to be most applicable (e.g. rain and snow types, temperature and pressure).

AR: As we responded to several comments above (see responses 2 and 7 in particular), although the 90th-percentile Λ profiles for rain and snow do not exactly match any of the theoretical formulations but they are close to certain combinations. However, the set of theoretical profiles that are close to the 90th-percentile profile do not appear to favour any rain or snow types or environmental conditions. Furthermore, one design goal for the new parameterization was for it to be applicable to CTMs that do not predict rain and snow types. This has been accomplished by working with an ensembles for rain and snow composed of all precipitation types. As for temperature and pressure effects, the sensitivity tests that were carried out (see response 4) showed that a wide range of temperature and pressure values only caused an uncertainty of at most 10% (with the exception of rain scavenging of particles in the Greenfield gap, for which the uncertainty increased to a maximum of 30%).

RC: (18) Conclusion, line 14, I am not convinced that closer agreement with field derived values can necessarily be termed ‘more realistic’. There are a variety of factors included in the field measurements that are not expected to be included in the theoretical formulations such that it is not clear whether a ‘more realistic’ agreement should be expected for the 90th percentile as opposed to any other percentile.

AR: We agree with the reviewer that closer agreement with field-derived values does not necessarily imply a more realistic theoretical treatment due to the various factors that are implicitly included in the field measurements but left out of the theoretical formulations. However, the situation here is that *all* of the theoretical values are “too far” from the field-derived values (e.g., Wang et al., 2010). Given this situation, those formulas that are closer to the field ones are considered to be more realistic.

Technical corrections:

AR: All of the technical corrections suggested by this reviewer have been made except the first one, for which we checked the values and found that the original one is correct.

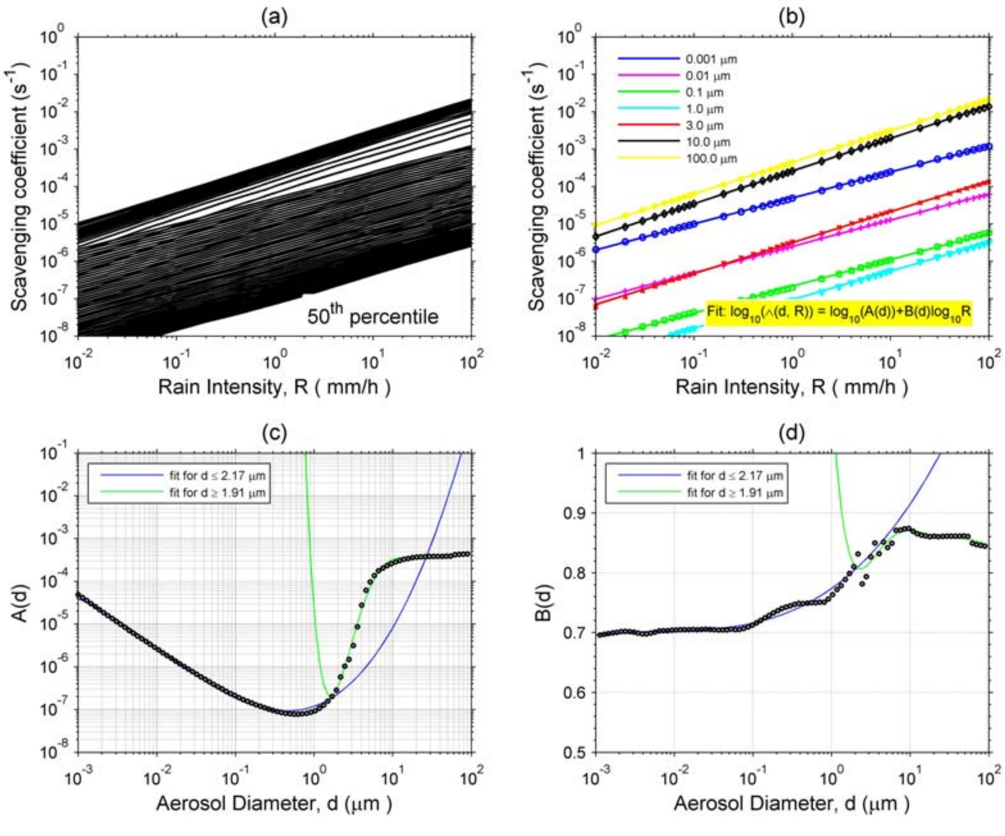

Fig. A. Same as in Fig. 2 for rain scavenging but based on 50th percentile

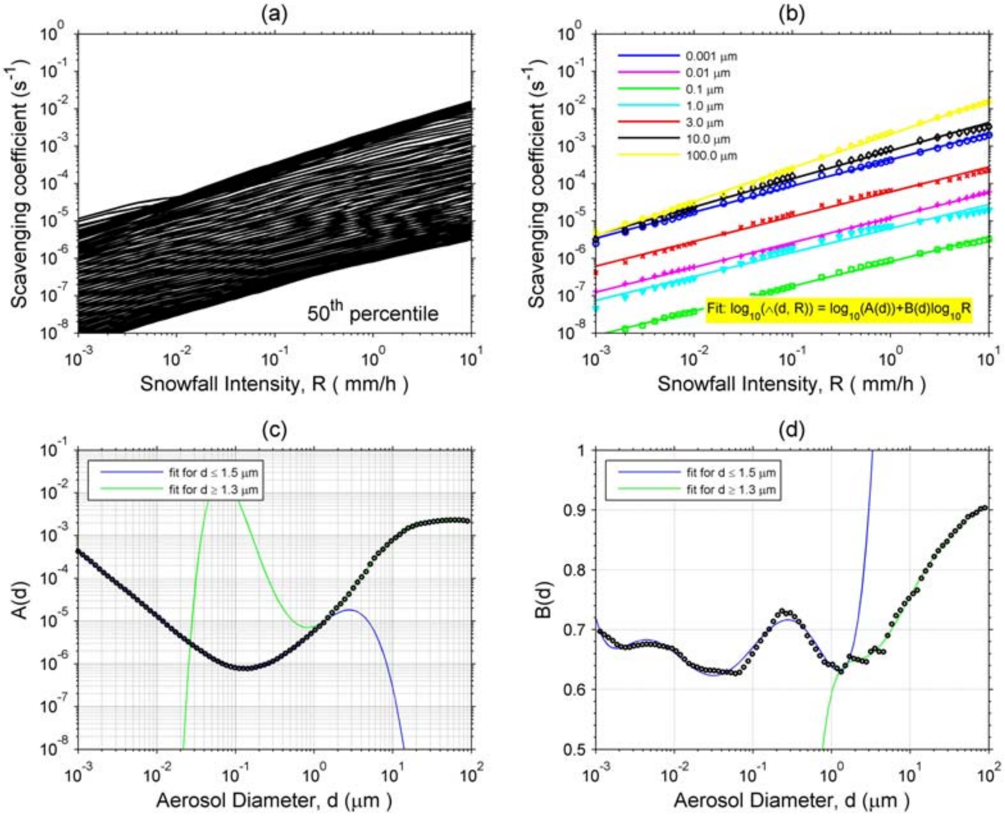

Fig. B. Same as in Fig. 5 for snow scavenging but based on 50th percentile

Reply to Reviewer #2

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's Responses

Specific Comments:

RC: (1) On pg. 5907, line 20 is written: "The ambient temperature was assumed to be 15 C for rain cases and −10 C for snow cases and the ambient pressure was assumed to be 1013.5 hPa. How these assumptions impact on the new parameterizations? I suggest to the authors to add these results in the MS.

AR: We have conducted sensitivity tests using different temperature and pressure values to address this comment. Temperature values of 5^{o}C and 30^{o}C for the rain cases and -5^{o}C and -30^{o}C for the snow cases and an ambient pressure value of 900 hPa for both the rain and snow cases were used in the sensitivity tests. The differences caused by using different ambient temperature and pressure values are generally within 10% of the previously reported results for all particle sizes for both rain and snow scavenging; the only exception is for rain scavenging of particles of sizes 0.1-2.0 μm, for which the bias can be up to 30%. We have presented these results in a new figure, Figure 8, and have added a summary of the results of these sensitivity tests and related discussions in the revised paper to a new section, Section 4.3.

RC: (2) On pg 5919, line 5 is stated that "The new parameterization . . . is more realistic than the majority of theoretical _(d) formulas". In order to support that I suggest to the authors to add a comparison with other parameterizations for both rain and snow in the Sect. 3.1 and 3.2.

AR: The development of this new parameterization is based on results from our three previous studies (Wang et al., 2010, 2011; Zhang et al., 2013), in which we conducted detailed comparisons of existing theoretical formulas with available field data and empirical formulas. A major conclusion and recommendation from our previous studies is that, as already stated in the Introduction of this paper, the upper range of available theoretical values should be used in chemical transport models. That is why we chose to use the 90th percentile of values from an ensemble data set calculated based on most existing theoretical formulas as the basis for developing the new scheme. The comparison of this new scheme with existing schemes and field data would be similar to the comparisons already presented in Wang et al. (2010) and Zhang et al. (2013), and we chose not to repeat what was already presented previously.

RC: (3) pg. 5902: Since “empirical” refers to something relying on or derived from observation or experiment, I suggest to the authors to change the title in “Theoretical development of new parameterizations for below-cloud scavenging...”

AR: Formulas generated from the fitting of data are usually called empirical formulas. The data themselves can be field-measured data or theoretically-produced data. The theoretical formulations for scavenging coefficient Λ have a semi-empirical aspect because the expressions for some product terms (e.g., terminal velocity) are based on empirical fits to measurements. In this study, the ensemble data set of scavenging coefficient values was first generated from theoretical (or semi-empirical) formulas and was then fitted to new formulas. Moreover, some of the choices in the calculation methodology such as the decision to consider 90^{th}-percentile values were based on consideration of measurements as well as theoretical values. We thus think it is appropriate to describe the new formulas as “semi-empirical” since they are neither purely theoretical nor purely empirical.

RC: (4) pg. 5904, line 10: The statement “the only exception is one controlled outdoor field experiment that obtained rain to a similar order of magnitude to the theoretical values.” has to be supported by the reference.

AR: The reference has been added in the revised paper.

RC: (5) pg. 5906, line5: “component parameters” are not appropriate terms. I suggest to use other terms all over the MS.

AR: We have changed the term “component parameters”, which was used to refer to the factors of the integrand product in Equation (2), to “product terms” throughout the text.

RC: (6) pg. 5907, line 5:“a number of size bins or sections”: the term “sections” is not usually used in aerosol microphysics, I suggest to be omitted.

AR: We believe the reviewer is referring to line 13, not line 5, of page 5907. The term “section” refers to one major modelling technique used in regional aerosol models to represent the continuous aerosol particle size distribution (i.e., a sectional representation). This terminology will be very familiar to AQ modellers even if it is not familiar to aerosol microphysicists. For example, here are a few references to papers that employ this term:

Jacobson, M.Z., 1997: Development and application of a new air pollution modeling system. Part II: Aerosol module structure and design. *Atmos. Environ.*, **31**, 131-144.

Meng, Z., D. Dabdub and J.H. Seinfeld, 1998: Size-resolved and chemically resolved model of atmospheric aerosol dynamics. *J. Geophys. Res.*, **103**, 3419-3435.

Seigneur, C., A.B. Hudischewskyj, J.H. Seinfeld, K.T. Whitby, E.R. Whitby, J.R. Brock and H.M. Barnes, 1986: Simulation of aerosol dynamics: a comparative review of mathematical models. *Aerosol Sci. Tech.*, **5**, 205-222.

Wexler, A.S., F.W. Lurmann and J.H. Seinfeld, 1994. Modelling urban and regional aerosols − I. Model development. *Atmos. Environ.*, **28**, 531-546.

We thus prefer to keep this term in the manuscript.

RC: (7) Fig. 1a and b: The caption should include what means red, black and yellow curves.

AR: The caption has been modified as recommended.