Reply to Reviewer 1

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

AC: We greatly appreciate this reviewer's constructive comments. We have carefully addressed all comments as detailed below, and we believe that the paper has been substantially improved as a result.

Major Points:

RC: Section 5 seems to be rather discontinuous with this earlier discussion. I would suggest modifications to Section 5 to indicate more clearly how the sensitivity of the scavenging coefficients to the factors discussed in Section 2 and 3 (and the abstract) relates to the predicted changes to the aerosol concentrations and distributions examined in Section 5. These changes to the predicted concentrations and distributions appear to be primarily related to the choice of parameterizations that are an empirical fit to field data, as opposed to theoretical calculations. The introduction should be strengthened to explain more clearly that the study will also consider parameterizations that are empirical fits to field data, in addition to the parameterizations based on theoretical calculations, which are the focus of Sections 2 and 3.

AC: The following major changes have been made in the revised paper to link the sections together better and to distinguish more clearly between theoretical and empirical formulations:

(1) In the second paragraph of 'Introduction' section, we now point out that both theoretical and empirical scavenging coefficient (Λ) formulas exist in the literature and that we review them together in this study. In the third paragraph of this section we state that Sections 2 and 3 focus only on existing theoretical Λ formulas and their associated input parameters whereas Sections 4 and 5 discuss both theoretical and empirical Λ formulas as well as field and controlled experiments. The revision of the Introduction provides a clearer outline of the paper so that readers can follow it more easily.

(2) The word 'theoretical' is added to the title of Section 3 and the words 'theoretical' and 'empirical' are added to the title of Section 4 to better reflect the focus of these sections.

(3) Substantial changes have been made in Section 5 to distinguish the results produced from theoretical and empirical Λ formulas. Other changes (e.g., adding a discussion of the results of sensitivity tests for the choice of integration time step) have also been made based on many specific comments provided by this reviewer.

(4) Revisions have also been made to Section 4 based on Comment 25 below from this reviewer to distinguish more clearly between field and controlled experiments as well as their agreements or disagreements with theoretical and empirical Λ formulas.

(5) The abstract has been revised to reflect revisions to Sections 4 and 5 (there was no need for changes for Sections 2 and 3).
(6) The Conclusions section has been revised with additional discussions based on several specific comments provided by this reviewer (e.g., Question 31 below).

Specific Points:

1) Page 2505, line 25 states that there have been no specific analytical expressions recommended. Section 4 starts with a reference to analytical formulas. Could the authors clarify this sentence in the Introduction?

AC: What we meant here is that there are a number of different analytical, empirical, or semi-empirical formulas available to describe raindrop size distribution, droplet terminal velocity, and raindrop-particle collection efficiency, all of which are needed for calculating scavenging coefficient (Λ). To date there has not been any community agreement or consensus as to which formula should be used for the above mentioned parameters in the calculation of Λ, which leads to different choices being made by different modelling groups, and to date no systematic uncertainty assessment had been done on the sensitivity of Λ to the choice of these input parameters. This is why in Section 3 the sensitivity of Λ to these input parameters was first considered, and why in Section 4 different combination of these parameters in the formulation of Λ found in the literature were then compared to each other and to measurements. Of course, in Section 4, empirically-derived formulations of Λ were also included.

In the revised paper, the second paragraph of the Introduction has been reorganized based on this comment and a few other comments provided by this reviewer: empirical and theoretical formulas of Λ were both mentioned; input parameters needed for theoretical Λ were then discussed; a wide variety of different formulas existing for these input parameters were pointed out; and the words 'analytical expression' have been removed since both empirical and analytical formulas co-exist.

2) Page 2508, line 10, and page 2509, line 6, refer to the Slinn (1983) equation as a "semi-empirical approximation", and an "analytical expression". Is this consistent wording?

AC: The Slinn (1983) formula for the raindrop-particle collection efficiency (E) was derived from dimensional analysis coupled with experimental data. In this sense, it can be called semi-empirical formula. To make the terminology consistent throughout the paper, we simply use the word 'formula' for all equations (e.g., those for Λ and for related parameters, including E). This is because many analytical formulas include parameters that are empirical or semi-empirical. The only exception is for Λ, for which we separate all formulas into theoretical ones and empirical ones, but for which all formulas are analytical expressions.

3) Could Figs. 1, 2, 4, and 6 be presented in Section 2? It might be helpful to see those plots during this earlier part of the discussion.

AC: This is a reasonable suggestion, but after careful consideration, we chose not to move these figures from Section 3 into Section 2. There were two reasons. First, Figures 1 and 2 show the contributions of different collection mechanisms to the collection efficiency (E). The calculations of E require knowledge of raindrop terminal velocity. If these figures were moved into Section 2.1, it might cause some confusion since the parameterizations for the raindrop terminal velocity are described in a later section. And second, Section 3 focuses on the sensitivity of Λ to a variety of formulations of raindrop-particle collection efficiency, raindrop number size distribution, and raindrop terminal velocity. Showing all figures together in this section seems to be the clearest way for the discussion to unfold.

4) Sections 2.1 and 3.1 discuss the Slinn parameterization for collection efficiency, and the alternative of a constant efficiency. How do these parameterizations compare to the collection efficiency expression given in Park et al. (2005), based on Jung and Lee (1998), and also the efficiencies used in Croft et al. (2009)?

AC: These two parameterizations, Park et al. (2005) and Croft et al. (2009), have been added in the revised paper (in Section 3.1 and in Figures 3, 8, 9 and 10) based on this comment. Λ values produced from these two parameterizations are smaller than Slinn's value for particles smaller than 0.2 μm but larger than Slinn's value for particles 2-10 μm. Detailed discussions on these two additional parameterizations have been added to the revised paper.

5) Page 2515, line 6, how do the results change with particle density?

AC: We have performed some sensitivity tests on the influence of particle density on raindrop-particle collection efficiency (e.g., assuming aerosol particles with a density of 1.77 g/cm^3 ($(NH_4)_2SO_4$) or 2.65 g/cm^3 (SiO_2)). The results show that the influence of aerosol density on raindrop-particle collection efficiency is negligible. This is because the density factor has a square-root dependence on aerosol density ($(\rho_p/\rho_w)^{1/2}$) and only affects inertial impaction. A brief discussion on this point has been added to the revised paper.

6) Page 2516, line 1, explains that smaller raindrops are more efficient collectors since they fall slower and there is more time for interactions with nearby particles. I thought that the definition of collection efficiency was time-independent – can you clarify this point?

AC: The definition of collection efficiency does not depend on time directly. However, E is a function of Reynolds number, which is in turn a function of terminal velocity. The smaller the terminal velocity, the smaller the Re, and thus the larger the E (as can be seen from Eq. 3).

7) Page 2516, line 12, quantify what is meant by 'significantly enhance'. In the abstract I think this is the one order of magnitude uncertainty in the scavenging coefficients attributed to collection efficiency, but I missed this in the Section 3.1 discussion. Also, I am still not sure if I am convinced whether this is truly an uncertainty, or just a difference related to a more complete, versus less complete description of the collection efficiency. Can you explain why you choose to call this an uncertainty?

AC: We agree with the reviewer that using the word 'uncertainty' might not be the best choice if only discussing Λ from Slinn's formula for E but with different collection mechanisms included or excluded. However, since this reviewer also suggested adding two other parameterizations, Park et al. (2005) and Croft et al. (2009), to the revised paper, it seems appropriate now to use 'uncertainty' to compare the different Λ formulations. So in the revised paper, at places where only Slinn's formula for E is mentioned (but with more or fewer collection mechanisms included), the word 'uncertainty' is avoided; but at other places where different formulas for E are compared, the word 'uncertainty' is still used. We also use quantitative measure in the revised paper when doing comparisons, e.g., 'with additional collection mechanisms included in the Slinn (1983)'s scheme, the predicted Λ can be increased by as much as one order of magnitude for particles 0.1-3 μm.

8) Figure 3, what causes the scavenging coefficients to increase for particles larger than 10 μm?

AC: A detailed investigation of this comment led to the finding that the increased Λ values for particles larger than 10 μm was caused by an error in the calculation of the interception term (hereafter referred to as $E_{int}(d_p, D_p)$). From Slinn's formula, it can be seen that this term is a function of the size ratio, d_p/D_p, of a particle and a droplet (a pair that makes collision). Generally, $E_{int}(d_p, D_p) << 1$ since $d_p << D_p$. However, in the case of $d_p/D_p > 1$, $E_{int}(d_p, D_p) > 1$ may occur. In this case, the formula of $E_{int}(d_p, D_p)$ should be a function of D_p/d_p instead of d_p/D_p. This is the reason for the unrealistic increase in scavenging coefficient in Fig. 3 for particles larger than D_p in size.

In the revised paper, we slightly modified Slinn's formula for $E_{int}(d_p, D_p)$. We replaced the size ratio of d_p/D_p in the formula by d_{small}/d_{large}, where d_{small} denotes the size of the smaller collision partner while $d_{l\arg e}$ denotes the size of the larger collision partner. This technique has been used in some large-scale models (e.g., Croft et al., 2009; Jacobson et al., 2003). We have re-calculated all of the model simulations with the revised formula and obtained more reasonable results. Note that all of the figures are re-plotted in the revised manuscript.

9) Page 2517, lines 9-12, quantify what is meant by ‘many fewer; and ‘not as large’ and ‘wider’.

AC: These qualitative words and phrases have been replaced with more quantitative measures in the revised paper. For example, this statement has been added: “By contrast, the gamma and lognormal distributions have fewer small droplets. For example, Table 2 shows that the percentages of droplets smaller than 0.1 mm for the DE gamma distribution and FL lognormal distribution are close to zero for all precipitation classes.” We also added the following sentence at the end of the paragraph: “However, the difference in N_{total} between DE and FL is smaller than a factor of 2.”

10) Page 2518, lines 20-21, quantify what is meant by ‘different representative drop diameters’. How much change in the representative diameter do you mean, and by how much does the scavenging coefficient change?

AC: Here we meant that different approaches are available in the literature for defining a ‘representative drop diameter’ to replace the whole droplet size spectrum. In order to reduce the computational burden, many large-scale aerosol transport models choose to use a monodisperse distribution by introducing a representative diameter D_r to replace the actual raindrop size distribution in the calculation of scavenging processes (e.g., Gong et al., 2003; Gong et al., 2006; Loosmore and Cederwall, 2004; Tost et al., 2006). This approach assumes that the scavenging coefficient calculated with D_r will match that computed from the full integral over all raindrop sizes (from Eq. (2) in the text). A power-law function has been commonly used to parameterize D_{r} ($=A{\cdot}R^{\mathrm{B}}$), where A and B are constants and change with the rainfall intensity R.

In Figure 5, we presented two Λ profiles derived from using two sets of different A and B values. One is used in AURAMS, where D_r is parameterized as $D_r = 0.7R^{0.25}$, which was originally proposed by Slinn (1977) and Slinn (1984) and determined based on the results given by Mason (1971) for fairly steady rains. The other is used in NARAC/LLNL, where $D_r = 0.97R^{0.158}$, which was generated from the median volume diameter for a gamma raindrop size distribution (Willis, 1984). Obviously, both D_r and Λ values change with the rainfall intensity. For example, for a rain intensity of 0.1 mm h^{-1} (Fig. 5a), the D_r difference is 0.28 mm (a factor of 1.7) and the corresponding Λ difference is about a factor of 5; for a rain intensity of 10 mm h^{-1}

(Fig. 5b), the D_r difference is 0.15 mm (a factor of 1.1) and the corresponding difference in Λ is smaller than a factor of 2. We have added a brief statement of the above conclusions to this paragraph.

11) Figure 5, what causes increased differences between the parameterizations for particles larger than 10 μm?

AC: See our answerer to Comment 8 above. The differences between different parameterizations no longer increased with size for particles larger than 10 μm once the code for the interception collision mechanism was corrected.

12) Page 2518, line 23, do you mean 'very close' to each other, or to the other parameterizations?

AC: The phrase "to each other" was added in this sentence in the revised paper.

13) Page 2519, line 15. A rainfall intensity of 1 mm/h was chosen. How do your results differ for more extreme rainfall intensities, such as 0.1 or 50 mm/h?

AC: We have done some sensitivity tests for different rainfall intensities. The impact of different $V(D_p)$ parameterizations on Λ values is generally small for all particle sizes. The largest uncertainty occurs for conditions of very weak rain (i.e., drizzle) due to the high concentrations of small droplets. In this case, Λ from using different $V(D_p)$ formulas is still within a factor of 2. As rainfall intensity increases, the differences in Λ decrease. The above results have been summarized and added to the revised paper.

14) Page 2520, line 25-26, quantify what is meant by 'very fast', 'quite fast' and 'slowest'.

AC: These qualitative words have been explained with real numbers in the revised paper. The sentences has been changed to: "Below-cloud scavenging is fastest for particles larger than a few microns in size, moderate for particles smaller than 0.01 μm in size, and slowest for particles in the range of 0.1-1 μm. For example, the observed Λ values were around $1\text{x}10^{-4}$ s^{-1} to $3\text{x}10^{-4}$ s^{-1} on average for particles in the diameter range of 3.5-10 μm (Volken and Schumann, 1993), around $1\text{x}10^{-4}$ s^{-1} for particles smaller than 0.01 μm (Davenport and Peters, 1978), and around $1\text{x}10^{-5}$ s^{-1} on average for particles in the range of 0.1-1 μm (Laakso et al., 2003)."

15) Page 2522, line 16, suggests that turbulence was also be an important process to consider. How does this relate to the work of Vohl et al. (2000)? I thought that turbulence may not be important for larger collectors such as raindrops.

AC: The reviewer is right in that ‘turbulence plays minor role for large droplets in the collection of particles’ based on the study of Vohl et al. (2000, 2001). For example, Vohl et al. (2001) found no enhancement of the collision efficiency for sub-micron particles collected by droplets with sizes between 346 μm and 2880 μm based on wind tunnel experiments. However, turbulence could play important roles due to the following two reasons:

(1) Under medium to light rain conditions, there are substantial numbers of rain droplets that do not belong to the large droplet size range (e.g., > 300 μm) for which turbulence plays no role. For example, Khain and Pinsky (1997) found that the collection efficiency due to turbulence was significantly enhanced for droplet collectors with size of 120 μm and for particles of size 4-6 μm.

(2) Turbulence in the atmospheric boundary layer causes vertical diffusion and thus transport of some particles (especially small ones) upwards into the cloud layer where they are then removed through in-cloud processes (Andronache et al., 2006).

We have added a brief discussion on the above two points to Section 4 of the revised paper and also discussed the second point in the Conclusions section.

16) Page 2523, line 12, suggests the need for new measurement data. Can you comment on whether this new data should be focused on laboratory or fieldwork? Since field observations inherently include all physical processes, do we need to be careful in deriving empirical parameterizations from field observations for use in large scale models that already include those additional physical processes separately?

AC: Laboratory data under controlled conditions will be very useful to verify the existing theoretical scavenging formulas (which only consider the collection process), especially considering that there is only such experimental one data set available for comparison at this time. Field data under different rain conditions and other environmental conditions are certainly also needed, considering the very small database on this topic.

The existing empirically-derived formulas might not be applicable to large-scale models where additional physical processes have already been included separately. The reviewer is right in that ‘more caution needs to be paid for deriving empirical parameterizations from field observations for use in large-scale models’. We have added the above discussion in the revised paper.

17) At the start of Section 5 there are few details about the model that you used. Could you provide a few additional details about the time-step, number of aerosol bins, and a general model description.

AC: No complex model was used to produce Figures 9 and 10. The results were simply generated from an integration of Equation 1 with very small time steps (10 s) and a large number of size bins (100) for both rain droplets (1 μm to 10 mm) and aerosol particles (0.001 μm to 100 μm) with a constant volume ratio between successive size bins. This information has been added to Section 5 in the revised paper.

18) Does the treatment of the aerosol size distribution introduce any further uncertainties? If a bin model is used, does the number of bins influence the predicted scavenging?

AC: The aerosol size bin resolution definitely has an impact on the predicted bulk and size-resolved aerosol concentration. $\Lambda(d_p)$ is the size-resolved scavenging coefficient expressing the removal rate of the aerosol particles between diameters d_p and $d_p + \Delta d_p$. Since the scavenging coefficient varies significantly with particle size, the use of a smaller number of size bins (and hence larger Δd_p) could result in smoothing for $\Lambda(d_p)$ values in the bin space. Several early studies (Gong et al., 2003; Foret et al., 2006) have conducted such sensitivity studies on both dry and wet deposition (note that both dry deposition velocity and the scavenging coefficient have 'V' shaped values with particle sizes) and these studies suggest that a size-bin of 12 or higher should produce reasonable results in aerosol transport models. In our study, we simply use a large number of size bin (100) to ensure accurate results.

19) Section 5 gives results for a rainfall intensity of 1 mm/hr. How would your results change for more extreme rainfall intensities, both lighter and heavier?

AC: We have conducted sensitivity tests for three different rainfall intensities: 0.1, 1, and 50 mm hr^{-1}. For the same total precipitation amount, 5 mm of rain, which is equivalent to 50 hr of rain at 0.1 mm hr^{-1}, 5 hr at 1 mm hr^{-1}, or 6 minutes at 50 mm hr^{-1}, the differences between different Λ (both theoretical and empirical) on the predicted aerosol number and mass concentrations are largest for the weakest rainfall intensity. The reasons for this behaviour can be explained by our answers to Comment 6 above. We have added some results and related discussion of these sensitivity tests in Section 5 of the revised paper.

20) The abstract states that predicted particle concentrations differ by more than a factor of ten, but I did not find this explicitly stated in Section 5. Please check this. Also, the factor of 10, or more, does not seem to be related to the uncertainties in the 3 dominant factors in the theoretical formulations of the scavenging coefficient. Rather, this seems to be related to choosing an empirical fit based on field measurements versus using theoretical formulas. I think that this source of the discrepancy should be more clearly pointed out in the abstract.

AC: The results in Section 5 show that the differences in the predicted particle concentrations due to the use of different theoretical Λ parameterizations can be smaller than a factor of 2 for particles with diameters less than 0.01 μm and larger than a factor of 10 for particles with diameters larger than 3 μm even after only a small amount of rain (e.g., 2-5 mm). However, the empirical Λ parameterizations can cause the differences in the predicted particle concentrations to be larger more than a factor of 10 for both very small particles and large particles (due to the much higher Λ values from the empirical formulas compared with the theoretical formulas). Both Section 5 and the Abstract have been revised significantly to distinguish between the results for theoretical and empirical Λ formulas.

21) Page 2524, line 27, can you give the primary cause for the largest differences that are found for ultrafine and coarse particles? Also quantify here what you mean by 'largest differences'.

AC: Λ values are highest for coarse particles and also very high for ultrafine particles. Thus, precipitation scavenging will have the largest impact on the concentration of coarse and ultrafine particles. This is the reason why the largest differences appear for these particle size ranges. The differences have now been explained and quantified in Section 5 of the revised paper.

22) Page 2525, line 4, could you check with your model whether the scavenging for 0.01 – 3 μm was important for long-lasting or very heavy rainfall? The second last sentence of the abstract seems to refer to this point, and also again in the conclusion. I think this should be examined further in this section if the result is in the abstract.

AC: As mentioned in the response to Comment 19 above, we have conducted additional sensitivity tests for three different rainfall intensities: 0.1, 1, and 50 mm hr^{-1}. To give a quantitative answer to this question, a new table (see below) has been added in the revised paper. The new table lists the percentage of particles removed for three particle sizes (0.01, 0.1, and 1 μm) after 5 mm of rain (i.e., 50 hr of rain at 0.1 mm hr^{-1}, 5 hr at 1 mm hr^{-1}, and 6 minutes at 50 mm hr^{-1}) by using two theoretical Λ formulas (the one with the highest Λ and the one with the lowest Λ from Figure 8) and one empirical Λ formula. As can be seen from this table, if based on a theoretical Λ formulation, a long-lasting rain or very heavy rain removes only a small fraction of particles in this size range (< 5%, but the actual fraction of particles removed depends on particle size, rain intensity, and total rainfall amount). However, if based on an empirical Λ (which is 1-2 orders of magnitude higher than a theoretical Λ), a long-lasting or a heavy rain can remove a very large fraction of particles in this size range (~10-70%). In the real world, the removal of particles from the atmosphere would be a coupled process involving droplet-particle collection, vertical transport (diffusion) of particles to low clouds and subsequent in-cloud removal, and many other processes (as reflected in the observed data that were used for deriving empirical Λ formulas). It can be concluded that precipitation scavenging can be significant for particles in the 0.01 – 3 μm size range, especially during long-lasting periods of precipitation.

Table: Predicted percentage removal for three particle sizes (0.01, 0.1 and 1 μm) using three Λ formulas after 5 mm of rain (50 hr at 0.1 mm hr^{-1}, 5 hr at 1 mm hr^{-1}, and 6 minutes at 50 mm hr^{-1}).

Rainfall Duration and Intensity	50 hr at 0.1 mm hr^{-1}			5 hr at 1 mm hr^{-1}			6 min at 50 mm hr^{-1}		
Λ Scheme \ Particle Size	0.01μm	0.1μm	1μm	0.01μm	0.1μm	1μm	0.01μm	0.1μm	1μm
Henzing et al. (2006)	7.1	0.7	0.3	3.4	0.3	0.1	1.0	0.1	0.04
Andronache et al. (2006)	32.2	5.1	4.6	13.1	1.9	1.8	2.6	0.4	0.4
Laakso et al. (2003)	100.0	72.2	91.6	78.4	17.2	30.5	60.5	10.9	20.0

23) How would results using the scavenging parameterization of Bae et al. (2006) compare to the results shown in Figures 9 and 10?

AC: Based on a moment method, Bae et al. (2006) proposed an analytical formula for scavenging coefficient as a function of collision efficiency, raindrop terminal velocity, and raindrop number size distribution. To obtain the analytical expression for Λ, two widely-used raindrop size distributions, Marshall-Palmer (Marshall and Palmer, 1948) and lognormal (Feingold and Levin, 1986), were assumed, the fall velocity formula was taken from Kessler (1969), and a simplified Slinn (1983) formula for collision efficiency was adopted. All related input parameters used by Bae et al. (2006) (i.e., raindrop spectra, raindrop terminal velocity, and raindrop-particle collision efficiency) have been discussed in our study. The only difference between Bae et al. (2006) and our study is related to numerics: Bae et al. (2006) used a moment method to derive the scavenging coefficient whereas we used numerical simulation with a sectional bin approach. For a sufficient number of size bins, the sectional method is direct, accurate, but computationally expensive whereas the moment method is computationally efficient but requires the use of complicated moment equations that have been derived based on series of assumptions and simplification for the related parameters (e.g., collection efficiency). In comparison with the results shown in Figs. 9 and 10 obtained with the sectional method, the results obtained by Bae et al. (2006) with the moment method are close to the results by Andronache (2003) shown in Figs. 9 and 10 when Bae et al. (2006) used the Marshall-Palmer raindrop number size distribution and close to the results by Mircea et al. (2000) when Bae et al. (2006) used the lognormal distribution of Feingold and Levin (1986). In conclusion, no significant difference was found between the results of Bae et al. (2006) and the results presented in this paper.

24) Page 2526, line 4-5, I think that the conclusion should point out that this underprediction related to field data was not seen in comparison with the in-door laboratory data, and should also mention that the additional known physical processes (transport, mixing, cloud and aerosol microphysics) that influence field data are not included in the theoretical calculations. This makes a fair comparison between field and theoretical calculations difficult.

AC: We agree with this comment. The Abstract, Section 5, and Conclusions have all been revised to distinguish between the theoretical and empirical Λ formulations and the comparison of their results with field and controlled experiments.

25) Page 2526, line 9-10. I am not sure that the findings can support the conclusion that 'new collection mechanisms need to be identified'. As you point out, the results in this article and previous work that you present (Andronache et al. (2006), Sparmacher (1993)) seem to suggest that models need to include additional physical processes to make a fair comparison with field data, as opposed to exposing deficits in the understanding of the below-cloud scavenging process itself.

AC: We agree with this comment. As noted in our response to the previous comment, the Abstract, Section 5, and Conclusions have all been revised to distinguish between the theoretical and empirical Λ formulations and the comparison of their results with field and controlled experiments. We think more numerical studies using comprehensive microphysics models should be conducted to quantify contributions from different processes to the total particle wet removal. Laboratory studies under controlled conditions are also recommended to provide data for further evaluations of the existing collection mechanisms. Section 5 and the Conclusions section have been revised to emphasize these recommendations and the recommendation to identify new collection mechanisms has been removed.

26) Page 2526, line 10, can you explain why turbulence might be important for collectors as large as raindrops?

AC: See our response to Comment 15 above.

27) Page 2526, line 14, what do you mean by 'overall below-cloud scavenging'? Do you mean the overall change to the aerosol distribution?

AC: We meant total scavenging, considering that the observed particle removal by precipitation not only includes droplet-particle collection but also includes contributions from other processes (e.g., vertical diffusion and subsequent removal). The word 'total' is added in a bracket.

28) Page 2526, line 23, can you explain what you mean by 'new collection mechanisms'? Is there some aspect of the impaction scavenging process itself that we are missing? Or do we need to better understand how the other various physical processes (transport, mixing, aerosol and cloud microphysics) additionally influence the aerosol concentrations and distributions?

AC: As in our response to Comment 25, instead of recommending the identification of new collection mechanisms, we think there is a compelling need to conduct theoretical and numerical studies using comprehensive microphysics models to quantify contributions from different processes to the total particle wet removal should be emphasized. We have modified the Conclusions section of the revised paper to emphasize this point and we have removed the recommendation concerning the identification of new collection mechanisms,.

29) Page 2527, last paragraph, this is an interesting point related to time-step. It seems that you have conducted some sensitivity tests. Would it be possible to include any of these results in an earlier section?

AC: We have added some discussion about the additional uncertainty in the predicted number and mass concentrations related to the integration time step to Section 5 of the revised manuscript.

30) A more general question, do you think that the term 'below-cloud scavenging' or 'impaction scavenging' is better suited to this process, since particle scavenging by rain could also occur in clouds?

AC: If only considering collection mechanisms, the term "impaction scavenging" is definitely a better description since it can be applied both inside and below clouds. However, most earlier aerosol transport models were only concerned about mass concentration and did not consider aerosol number concentration. Thus, for aerosol mass, in-cloud impaction scavenging will not be an important process since most aerosol mass will already have been removed by nucleation scavenging. From this perspective, any distinction between impaction scavenging and below-cloud scavenging by rain will be minor. However, with the development of size-resolved aerosol transport models, there is greater interest in the explicit simulation of aerosol number concentration. In this case, a generalization to impaction scavenging will be required.

As discussed in Zhang and Vet (2006), the mechanisms of impaction scavenging inside a cloud and below a cloud are basically the same. The only difference is that cloud droplets inside a cloud are generally much smaller and more numerous than falling raindrops below the cloud base. Thus, careful parameterization of the collection efficiency of small aerosol particles by small droplets is very important for representing in-cloud impaction scavenging. For example, in-cloud impaction scavenging needs to consider the contribution of turbulence (turbulent inertial motion and turbulent shear) to the total collection efficiency whereas below-cloud impaction scavenging might neglect this effect.

In the present study, we have only mentioned the term 'below-cloud scavenging' since the measurements used for comparison were conducted below cloud.

31) Based on their work, could the authors make any recommendations in regard to a preferred approach for the representation of below-cloud particle scavenging in large-scale models?

AC: From the results shown in this study, we could make the following hypothesis: 'the values shown in Figure 8 from the majority of field measurements were conducted at heights close to the surface where vertical diffusion and turbulence were strongest; these physical processes could have substantially enhanced particle removal by transporting some particles upwards where they can then be scavenged by microphysical processes inside low clouds. These additional processes may be the main cause of the large discrepancy between the theoretical and the field-derived values seen in Figure 8'. Evidence supporting this hypothesis is listed in our reply to reviewer 2.

If the above hypothesis is true (and we believe that to be likely), then the empirical formulas for Λ seem unsuited to aerosol transport models in which many additional processes that directly or indirectly contribute to the particle removal have already been considered separately.

Since the current theoretical Λ formulas can produce comparable Λ values to those derived from controlled experiments (where addition processes contributing to particle removal were limited), they seem to be reasonable choices for use in aerosol transport models. To construct a theoretical Λ parameterization for aerosol transport models, we would offer three recommendations:

(1) Choose a theoretical Λ formula that produce highest Λ for particles of 0.01-3 μm. This is because even the highest Λ produced from theoretical formulas are still more than one order of magnitude smaller than the 'non-controlled field measurements' (although slightly higher than the 'controlled experiment', which should be reasonable considering that there must be some processes that are not considered separately in aerosol transport models but that contribute to the particle scavenging process). This requires using the highest theoretical collection efficiency, E, which argues for inclusion of all known collection mechanisms in

the framework of Slinn (1983)'s Λ formulation.

(2) Although a full droplet spectrum (the gamma function of de Wolf, 2001 is proffered) is the best choice, the use of a representative drop size in aerosol transport models should also be acceptable given that the uncertainties from such an approach are of a similar order of magnitude to those from other known or unknown processes (see also the response to Comment 10). However, if a full droplet spectrum is used, the theoretical formula of Beard (1976) is recommended for use in the calculation of raindrop terminal velocities since this formula provides a good estimate over the full range of raindrop sizes.

(3) If in-cloud impaction scavenging is included, different droplet spectra (or representative particle size) from those used for below-cloud scavenging need to be used since small droplets dominate within clouds. Also, the contribution of turbulence to the collection efficiency needs to be considered.

The above discussions have been added in Conclusions section in the revised paper.

32) Table 2: explain acronyms MP, DE, and FL.
AC: Done.

33) Figure 5: explain acronyms MP, DE, and FL.
AC: Done.

34) Technical correction: 1) Page 2524, line 26, 'precipitations' → 'precipitation'
AC: Done.

References mentioned above

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