Reply to Reviewer 2

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

AC: We greatly appreciate this reviewer's constructive comments. We have addressed all comments in the revised paper as detailed below.

Specific comment:

RC: As the authors conclude, field scavenging measurement data is biased by aerosol processes like condensation and coagulation and in addition, especially turbulence could enhance the value of collection efficiency between the raindrop and aerosol particles significantly. Therefore, the comparison of existing theoretical studies and measurements is somewhat difficult. For example in the case of the urban aerosol distribution (Fig. 10), it could be estimated that how many particles are removed for example by the coagulation compared to that of scavenging during the same time period?

AC: We agree with this comment and we are conducting further detailed studies that will be presented in a separate paper. As noted by this reviewer and also by Reviewer 1 (Question 24), there are additional known physical processes (transport, mixing, cloud and aerosol microphysics) that influence field data that are not included in the theoretical calculations. Thus, our next study will focus on quantifying the contributions of these and other processes to the overall (total) scavenging coefficient (Λ) using a comprehensive cloud microphysics model and the approach described in Zhang et al. (2004). The major goal of this detailed study will be to evaluate the following hypothesis: the Λ 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 the particle removal by transporting the particles into upper layers where they were then scavenged by microphysical processes (Andronache et al., 2006). This is expected to be the main cause of the large discrepancy between the theoretical Λ and the field-derived Λ values as seen in Figure 8.

There are several lines of evidence to support the above hypothesis: (1) The Λ values obtained through a controlled experiment, where vertical diffusion and turbulence were minimum (Sparmacher et al., 1993), were much lower than all of the other field measurements but agreed quite well with theoretical Λ values (Figure 8); (2) It is expected that vertical diffusion and turbulence should have a big impact on raindrop scavenging of small particles but much less impact on large particles. This also explains why the theoretical Λ values agree quite well with most field measurements for particles larger than 3 μm but are one to two orders of magnitude smaller for small particles when compared to field measurements (except the controlled experiment of Sparmacher et al., 1993) (Figure 8); (3) Using a simplified scavenging model that includes mixing of ultrafine particles from the boundary layer into cloud, Andronache et al. (2006) found good agreement of the overall particle scavenging when compared to the field data of Laakso et al. (2003); (4) The Λ values shown in Figure 8 of Zhang et al. (2004) produced from a detailed cloud microphysics model (where the droplet-particle collection mechanisms were similar to those theoretical formulas shown in Figure 8, but where the vertical diffusion process was also considered) also seem to be much higher than the simple theoretical Λ values shown in the present study.

All the above evidence suggest that the measured Λ not only includes contributions from the theoretically-defined collection processes but also includes contributions from many other physical processes. A set of sensitivity tests will be designed for different aerosol distributions, different precipitation rates, and different vertical diffusion scenarios using the model approach described in Zhang et al. (2004). These results cannot be included in this paper, but more detailed discussion on this point has been added in Section 4 of the revised paper to address this comment.

Technical comments:

RC: Page 2504, line 22: "The differences for submicron-sized particles. . ." The word submicron is used throughout the paper to describe the particles between 0.01-1 μm (particles in Aitken and accumulation mode). However, literally submicron refers to all particles below 1 μm. To avoid confusion I would give a size range when it is not all particles < 1 μm in question.

AC: The particle size or particle-size range is now explicitly stated wherever particle size is mentioned in the revised paper.

RC: Page 2508, line 20: For some reason the table numbered as 4 is used for nomenclature?

AC: Nomenclature is listed as Appendix B in the revised paper.

RC: Page 2510, lines 16-17: "α is an empirical parameter that can vary between 0, which corresponds to neutral particles and 7, which corresponds to highly electrified clouds". This is a bit confusing sentence because α exists both in raindrop and particle charge equations. At the same time, is it possible to use different value for α to raindrops and particles? If yes, for instance the symbols α p and α r could be used.

AC: α is an empirical parameter that depends only on the electrical environment of clouds (i.e., neutral, weak, or highly electrified clouds). The mean charges on raindrop (Q_r) and on aerosol particle (q_p) were expressed as a function of their sizes, separately. Note that the expression of the electrostatic collection efficiency (Eq. 6) is from Andronache (2004) and Andronache et al. (2006).

RC: Page 2512, line 6: ". . .seem to be better in. . ." Is it better or not and how this conclusion was made?

AC: The classical two-parameter exponential distribution (MP) was determined by Marshall and Palmer (1948) by fitting the observed data of Laws and Parsons (1943). However, De Wolf (2001) found that a gamma function fits better than MP's exponential function at the small end of the droplet range (R<0.5–1 mm) when compared with the same data set of Laws and Parsons (1943). The exponential function (see Eq. 8) includes a fixed intercept parameter N_{0e} that predicts maximum droplet number concentration for droplets with sizes approaching zero diameters. However, many electromechanical disdrometer observations and theoretical studies have shown that the intercept parameter is far from constant and systematically depends on precipitation type, rain intensity, and stage of development (e.g., Waldvogel, 1974; Sauvageot and Lacaux ,1995; Zhang et al., 2008). To improve the characterization of raindrop size distribution over the exponential distribution, three-parameter gamma distributions (or normalized gamma distributions) (see Eq. 9 in manuscript) have been proposed and widely used (e.g., Ulbrich, 1983; Willis, 1984; Willis and Tattelman, 1989; Tokay et al. 2001; Zhang et al. 2001, 2003; Bringi et al. 2002, 2003; Brandes et al. 2004a, b; Henzing et al., 2006). The results have shown that gamma distributions are usually better at representing the characteristics of observed raindrop size distributions at the small raindrop end than exponential functions. Besides the gamma functions, Mircea et al. (2000) proposed that raindrop size distributions could also be well described by lognormal functions on the basis of analyzing the long-time measurements of raindrop size spectra in the Mediterranean area: Israel (Feingold and Levin, 1986) and Spain (Cerro et al., 1997).

Thus, we concluded that the gamma and lognormal distributions are better able to represent the raindrop size distribution than is the MP exponential distribution. A brief discussion based on the above explanation has been added in the revised paper.

RC: Page 2512, line 7: ". . .small-particle end. . ." should probably be small raindrop end.
AC: Corrected.

RC: Page 2515, line 3: Ultrafine particle size range is not determined until here although it has been used already earlier.
AC: The size range of ultrafine, submicron and coarse particles are now explicitly defined in the revised paper wherever particle sizes are mentioned.

RC: Page 2515, lines 4-5: ". . .Stokes number St. . ." and ". . .Stokes number St*. . ." Parenthesis around St and St* are missing.
AC: Corrected.

RC: Page 2523, lines 8-10: ". . .this parameterization is valid only for particles 0.01-0.5μm. . .". However, the next sentence says: "Fig. 8 suggests that this parameterization overestimates Λ values for < 0.01 μm and > 10 μm particles". This is trivial, because they just told that parameterization shouldn't be used outside the given range. Also, that should be noticed in Figure 8.
AC: A note has been added to Fig. 8 in the revised paper. This sentence has also been removed from the text.

RC: Page 2525, line 26: smaller instead of "small".
AC: Corrected.

RC: Page 2530, line 19: full stop is missing in the end of the sentence.
AC: Corrected

RC: Table 3: Explain "Types" in the caption.
AC: Explanation added.

RC: Figures in general: in print, it is difficult to distinguish different colors. Also different line types or symbols should be used.
AC: We have tried to use different colours, line types, and some symbols in the revised paper to improve the figures.

RC: Figure 4, in caption: "MP and DE represent stratiform rain, JD drizzle rain, ..." If these parameterizations are meant for different rain types, they should only be compared inside the rain type in question.
AC: We agree that, ideally, the size distribution for different rain types should only be compared within the particular rain types. However, there are only very limited empirical formulas available. Besides, even the same rain type could have substantially different droplet spectra due to different precipitation rate, different rain formation processes (e.g., different initial CCN), etc. Considering that the main purpose of the present study is to compare the shape of different empirical spectra that can be used for calculating scavenging coefficient, we simply combined them. We have modified figure caption to avoid the words 'drizzle' and 'storm' to reflect better the main purpose of this figure.

RC: Fig. 9: Y-axis could be normalized to for example 100 or 1.
AC: Fig. 9 has been modified accordingly.

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