Response to Reviewer #1

Overall, this manuscript makes a nice contribution to understanding observations of below-cloud wet scavenging and the need to remove air-mass changes. It deserves to be published. However, I have some comments that I'd like to be addressed. I'm particularly concerned about Figure 8 and its interpretation, and this is the focus of my last 4 comments.

Response: Thanks for providing helpful and constructive comments, which help us further improve the manuscript. The changes in the revised manuscript were highlighted in red color. Below is our point-by-point response to each comment.

Comment 1:

The manuscript needs a general review of English (including title and throughout). Generally, the issues don't prevent the points from being understood, but things can be cleaned up to make the manuscript a bit easier to read. As an example with the title, it would be more common to say, "The variation of the particle number size distribution during the rainfall: wet scavenging and air masses changing".

Response: Thanks for the comment and suggestion. We have conducted a general review of English through our co-authors, which has made the manuscript more readable. Please see the revised version for details.

Comment 2:

Section 2.2: The criteria listed here have somewhat arbitrarily chosen cutpoints. How sensitive are the results to changes in these cutpoints?

Response: Thanks for the comment. We reviewed the literatures related to below-cloud wet scavenging studies based on field measurements. In the articles that specifically describe the selection criteria for rainfall events (Blanco-Alegre et al., 2018;Cugerone et al., 2018;Geng et al., 2019;Laakso et al., 2003;Luan et al., 2019;Pryor et al., 2016;Roy et al., 2019;Wang et al., 2014), there is no uniform standard in the selection criteria. The relevant cutpoints in this study were chosen based on the existing cutpoints in the literature mentioned above.

To test the sensitivity of the cutpoints to the results, we have reduced all cutpoints of the meteorological selection criteria by 20%, which makes the screening condition stricter. The strict meteorological selection criteria is: (i) the change in temperature at any adjacent hour during the rainfall events no greater than 4.8 ℃, (ii) the change in RH at any adjacent hour during the rainfall events no greater than 16%, (iii) the wind speed less than 3.2 m s^{-1} and (iv) the change in wind direction no more than 72° at the start and end of the rainfall event. The number of events inevitably decrease (normal events from 122 to 91, high BWSC events from 48 to 30). Nevertheless, the changes in BWSC after using strict cutpoints is not significant compared to the previous one (Fig. R1). Therefore, the chosen cutpoints don't affect the major results of this study. Considering the referee's comments, we have added the discussions on the selection of cutpoints in section 2.2. Please see Line 131-132 and Line 142-143.

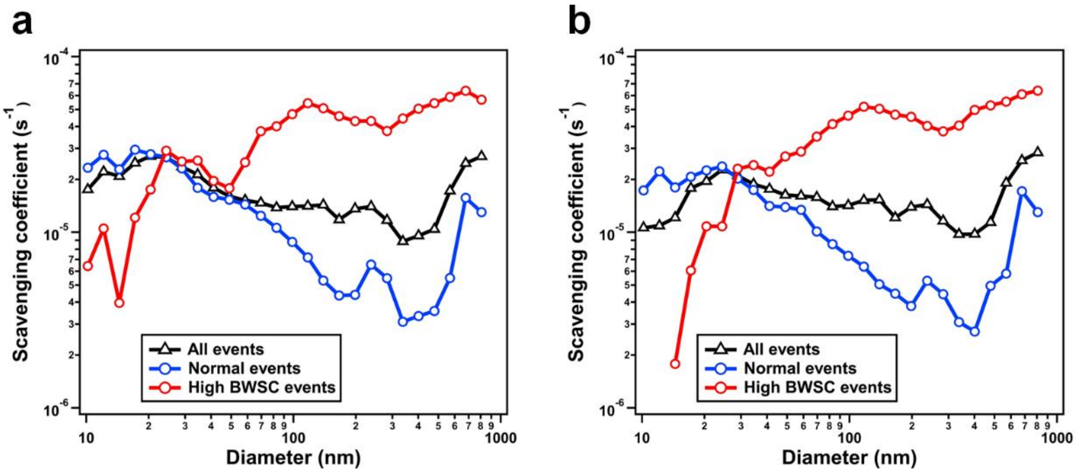

Figure R1. Below-cloud wet scavenging coefficients at the SORPES station for the (a) strict meteorological selection and (b) original selection criteria.

Comment 3:

Figure 2: I found it hard to see the "theory" curve on the plot (the pink was very light), please make it darker or make it a line.

Response: Thanks for the comment and suggestion. We have made the "theory" curve of the Fig. 2 darker in the revised version. Please see Figure 2.

Comment 4:

Figure 2: What assumptions were made for the "theory" curve? The rainfall rate and the rain size distribution will affect this curve. Related, the rain conditions for each of the obs studies on the plot may different, so it's worth discussing these potential differences.

Response: Thanks for the comment and suggestion. The assumptions of the "theory" curve are from Andronache et al. (2006). Raindrop-particle collection efficiency is from Slinn (1983) with the phoresis and electric forces taken into account; raindrop number size distribution is Marshall-Palmer distribution; raindrop terminal velocity is from Atlas and Ulbrich (1977); rainfall intensity is 10 mm/h. The rainfall rate and the rain size distribution do affect the "theory" curve. However, the differences between the curves obtained from different theoretical calculation is relatively small compared to their differences with field measurements (Wang et al., 2010). The rain conditions for each previous observation studies are different. In general, the stronger rainfall intensity lead to higher observed scavenging coefficient.

Considering the referee's comments, we have added more descriptions of the assumptions of the "theory" curve in the revised manuscript. Please see Line 171-173. And the relevant discussions on the rain conditions in each observation study are added to the revised manuscript. Please see Line 181-182 and Table1.

Comment 5:

L241-242: I don't understand this statement. It might be because I'm not sure what a "backwards air mass" is (do you mean "back trajectory"?).

Response: Thanks for the comment. The word "backward air mass" is not clear enough here and can be misunderstood. We have replaced "backward air mass" with "backward trajectory" in the revised version. Please see Line 253.

Comment 6:

Figure 6: Is there a reason why type iv is not plotted here? Should mention why.

Response: Thanks for the comment and suggestion. The type iv's backward trajectories have no clear horizontal rotation or vertical height change (Fig. 5d). Therefore, we didn't add the type iv to Fig. 6. Considering the referee's comments, we have added the description in the revised manuscript. Please see Line 296-298.

Comment 7:

Figure 7: The CO change here seems like the most straightforward evidence of the airmass change, given that it's a non-scavenged species. Not necessary, but this could be made more prominent in the paper (e.g., abstract, moving it forward in the results, etc.).

Response: Thanks for the suggestion. We have emphasized the characteristic of the CO change in the abstract and conclusion. Please see Line 19-20 and 386-388.

Comment 8:

Figure 8: What size are these BWSC coefficients for? Is it for the total number of particles, regardless of their size? If true, this complicates the interpretation of the figure since there is likely a correlation between the particle number concentration and the size of the particles. I recommend instead making this plot for the BWSC for a specific size, which makes for a fairer comparison (or at least having this as a second panel).

Response: Thanks for the comment and suggestion. The BWSCs in Fig. 8 are the median over particle size range from 10 nm to 500 nm. In the revised manuscript, we presented the BWSCs for each size bin (Fig. R2c, d).

As shown in Fig. R2c, the dependence of BWSC on rainfall intensity for particles below 100 nm (i.e. ultrafine particles) is similar with that of total particles, since the ultrafine particles dominate the particle number concentration at SORPES. The BWSCs are low for the particles larger than 100 nm due to the exist of "Greenfield" gap (Greenfield, 1957). Nevertheless, the increase in BWSC with increasing rainfall intensity can also be found for the particles larger than 100 nm. The dependence of BWSC on particle number concentration prior to rainfall event is obvious for ultrafine particles as well (Fig. R2d). In general, although the BWSCs are related to the particle size, the dependences of BWSCs on rainfall intensity as well as particle number concentration is uniform over all the size bins.

Considering the referee's comments, we have added the BWSCs in each size bin in Fig. 8 and the discussions of BWSCs for different size bins in the revised version. Please see Figure 8 and Line 357-365.

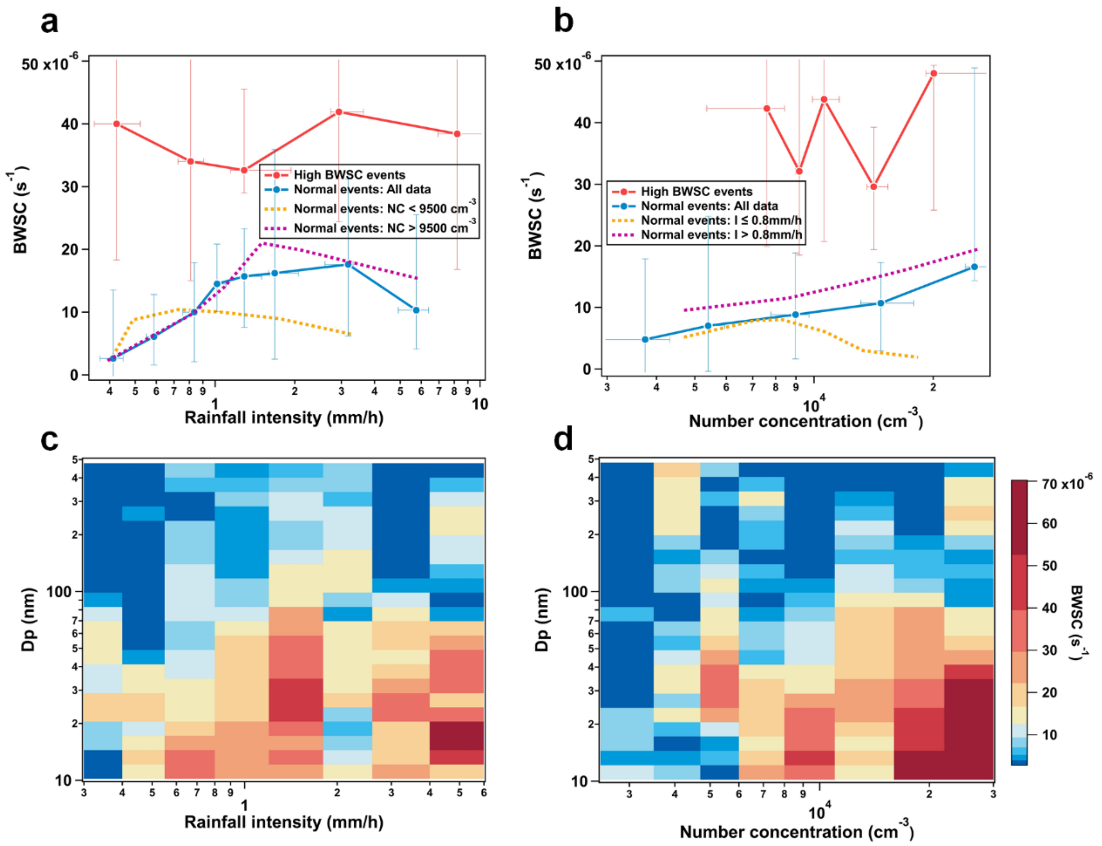

Figure R2. Below-cloud wet scavenging coefficients (median over particle sizes 10-500 nm) for high BWSC events and normal events as the function of (a) rainfall intensity and (b) particle number concentration prior to rainfall events. The dots represent the median and the error bars represent the upper and lower quartiles. Below-cloud wet scavenging coefficients in each size bin for normal events as the function of (c) rainfall intensity and (d) particle number concentration prior to rainfall events.

Comment 9:

L327-329: It does not make physical sense that the BWSC depends on number concentration. Below-cloud wet scavenging is a 1st-order loss process, particles should not be influencing other particles' ability to be scavenged. It seems much more likely that particle number and the average size of the particles are at least somewhat correlated, and that is driving the relationship here. This is why making Figure 8 show the BWSC for a specific size would be easier to interpret. (Also, is there a relationship between rain rate or the rain drop size distribution and the number of particles? A correlation here seems less likely than the relationship between particle number and particle size, but worth checking since it could also influence the interpretation of Figure 8.)

Response: Thanks for the comment and suggestion. As shown in Fig. R2d, the BWSCs increase with the increasing particle number concentration prior to rainfall event in most of size bins. Theoretically, below-cloud wet scavenging is a 1st-order loss process and should not have the relationship with the particle number concentration prior to rainfall event. The basic equation of variation in the particle number concentration $c(d_p)$

due to rainfall scavenging is described by Eq. (1) (Seinfeld and Pandis, 2016):

$$\frac{dc}{dt} = -\lambda c \quad (1)$$

λ is the scavenging coefficient (i.e. BWSC). As shown in Fig. R3a, the particle number concentration varies exponentially with time if no other processes (other sources or sinks of particles) are present during the rainfall event. The slope of the line is the scavenging coefficient (λ) and does not vary with the particle number concentration prior to rainfall (c_0).

However, in the real ambient environments, other processes such as particle formation, primary emissions, coagulation, etc. cannot be excluded. For example, if there is a stable source of particles, the variation of particle number concentration can be described by:

$$\frac{dc}{dt} = -\lambda c + A. \quad (2)$$

A is the formation rate of the particles due to the stable source. The variation of particle number concentration based on Eq. (2) is shown in Fig. R3b by assuming consistent BWSC and formation rate.

In field measurements, it is common to use Eq. (3) to calculate the BWSC:

$$\lambda = -\frac{1}{t_1 - t_0} \ln\left(\frac{c_1}{c_0}\right). \quad (3)$$

c_0 and c_1 are the median particle number concentrations before (t_0) and after (t_1) the rainfall event, respectively. Assuming same duration of rainfall event, the BWSCs can be calculated based on Eq. (3) for different initial particle number concentration (dash lines in Fig. R5b). Although the actual BWSC is consistent, the calculated BWSC can increase with the increasing initial particle number concentration. Therefore, the relationship between BWSC and particle number concentration prior to the rainfall event can be caused by other processes except for below cloud wet scavenging. In real ambient environments, other processes perturb the calculation of BWSC when using the BWSC calculation method as described by Eq. (3). We thereby highlight the need for further research on BWSC calculation methods based on field measurements.

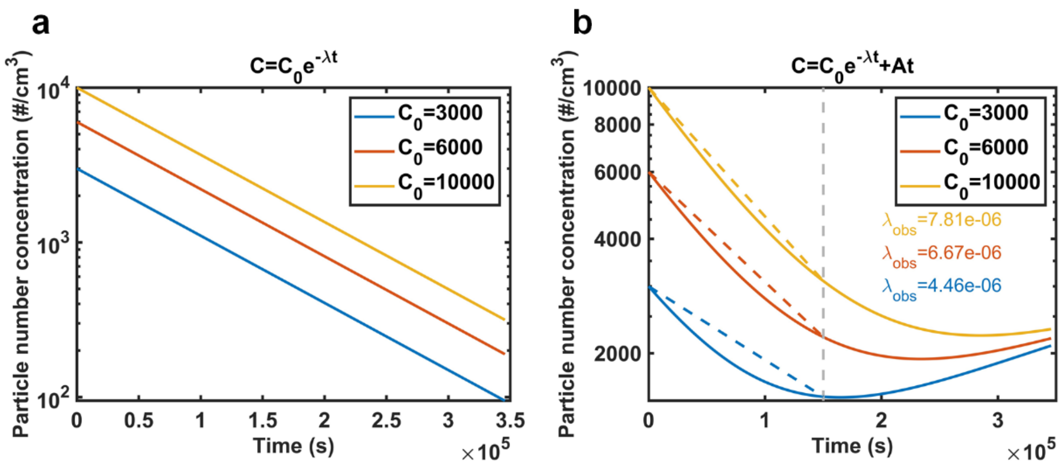

Figure R3. The variation of particle number concentration with time during rainfall under theoretical conditions (a) without and (b) with the stable source. (Assuming the scavenging coefficient (λ) of 1×10^{-5} s^{-1} and the particle formation rate (A) of 5.8×10^{-3} cm^{-3} s^{-1})

We checked the relationship between rainfall intensity and particle number concentration and they were not significantly related.

Considering the referee's comments, in the revised version, we have modified the discussions on the dependence of BWSCs on particle number concentrations prior to rainfall events in Line 366-379 and Figure S16.

Comment 10:
L341-342: How? Below-cloud wet scavenging is a first-order process. It should not depend on the number. However, it does depend on particle size. See the two comments above. The interpretation here seems incorrect.
Response: Thanks for the comment. Based on the two comments and responses above, we have modified the relevant discussions in section 3.4. Please see Line 357-379.

Comment 11:
L342-344: Unless the particle concentrations are extremely high, below-cloud scavenging should be removing particle number much faster than coagulation.
Response: Thanks for the comment. Based on the comments 8-9 and responses, we have modified the relevant discussions in section 3.4. Please see Line 357-379.

References:

Andronache, C., Grönholm, T., Laakso, L., Phillips, V., and Venäläinen, A.: Scavenging of ultrafine particles by rainfall at a boreal site: observations and model estimations, Atmos. Chem. Phys., 6, 4739-4754, 10.5194/acp-6-4739-2006, 2006.

Atlas, D., and Ulbrich, C. W.: Path- and Area-Integrated Rainfall Measurement by Microwave Attenuation in the 1–3 cm Band, Journal of Applied Meteorology and Climatology, 16, 1322-1331, 1977.

Blanco-Alegre, C., Castro, A., Calvo, A. I., Oduber, F., Alonso-Blanco, E., Fernández-González, D., Valencia-Barrera, R. M., Vega-Maray, A. M., and Fraile, R.: Below-cloud scavenging of fine and coarse aerosol particles by rain: The role of raindrop size, Quarterly Journal of the Royal Meteorological Society, 144, 2715-2726, 10.1002/qj.3399, 2018.

Cugerone, K., De Michele, C., Ghezzi, A., and Gianelle, V.: Aerosol removal due to precipitation and wind forcings in Milan urban area, Journal of Hydrology, 556, 1256-1262, 10.1016/j.jhydrol.2017.06.033, 2018.

Geng, T., Tong, H., Zhao, X., and Zhu, Y.: Effect of Wet Deposition on the Removal Efficiency of Particulate Matter in the Yangtze-Huaihe Region, Research of Environmental Sciences, 32, 273-283, 2019.

Greenfield, S.: Rain scavenging of radioactive particulate matter from the atmosphere, Journal of Meteorology, 14, 115-125, 1957.

Laakso, L., Grönholm, T., Rannik, Ü., Kosmale, M., Fiedler, V., Vehkamäki, H., and Kulmala, M.: Ultrafine particle scavenging coefficients calculated from 6 years field measurements, Atmospheric Environment, 37, 3605-3613, 10.1016/S1352-2310(03)00326-1, 2003.

Luan, T., Guo, X., Zhang, T., and Guo, L.: The Scavenging Process and Physical Removing Mechanism of Pollutant Aerosols by Different Precipitation Intensities, Journal of Applied Meteorolgical Science, 30, 279-291, 2019.

Pryor, S. C., Joerger, V. M., and Sullivan, R. C.: Empirical estimates of size-resolved precipitation scavenging coefficients for ultrafine particles, Atmospheric Environment, 143, 133-138, 10.1016/j.atmosenv.2016.08.036, 2016.

Roy, A., Chatterjee, A., Ghosh, A., Das, S. K., Ghosh, S. K., and Raha, S.: Below-cloud scavenging of size-segregated aerosols and its effect on rainwater acidity and nutrient deposition: A long-term (2009–2018) and real-time observation over eastern Himalaya, Science of The Total Environment, 674, 223-233, 10.1016/j.scitotenv.2019.04.165, 2019.

Seinfeld, J. H., and Pandis, S. N.: Atmospheric chemistry and physics: from air pollution to climate change, John Wiley & Sons, 2016.

Slinn, W. G. N.: Precipitation scavenging, Atmospheric Sciences and Power Production – 1979, chap. 11, 1983.

Wang, X., Zhang, L., and Moran, M. D.: Uncertainty assessment of current size-resolved parameterizations for below-cloud particle scavenging by rain, Atmos. Chem. Phys., 10, 5685-5705, 10.5194/acp-10-5685-2010, 2010.

Wang, Y., Zhu, B., Kang, H., Gao, J., Jiang, Q., and Liu, X.: Theoretical and observational study on below-cloud rain scavenging of aerosol particles, Journal of University of Chinese Academy of Sciences, 31, 306-313, 2014.