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Atmospheric
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Discussions

Interactive comment on "On the discrepancies between theoretical and measured below-cloud particle scavenging coefficients for rain – a numerical study" *by* X. Wang et al.

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We appreciate the reviewer's comments, which improved the clarity of the paper. We have addressed the reviewer's general and specific comments and we have revised the paper as detailed below.

RC- Review Comments; AC – Authors' Comments

General Comments

RC: Authors explain the discrepancies between theoretical and measured below-cloud particle scavenging coefficients for rain by the contribution of vertical diffusion for

aerosol particles of diameter >0.01μm. However, in the conclusion, authors claim the significant contribution of vertical diffusion to overall size-resolved scavenging coefficient for submicron particles of diameter <0.005μm is only under the conditions of "weak precipitation" or drizzle (0.1mmh-1 and 1.0 mmh-1).

AC: We focused our discussion in the main body of the paper on particles >0.01 μm. This is because the contribution of vertical diffusion to the overall scavenging is largest for particles with sizes of 0.01- 2 μm. For particles smaller than 0.01 μm, the contribution from vertical diffusion decreases with the decrease of particle size and is only visible under weak precipitation conditions. The statement in the Conclusions section that the reviewer referred to is an addition to the major discussion presented in the main text, that is, that "largest effects were found for particles 0.01-2 μm under all precipitation intensities and small effects could also be detected for very small particles (e.g., <0.005 μm) under weak precipitation conditions". We have added some discussion for particles smaller than 0.01 μm to the main text and we have also revised the Conclusions to make the statement more straightforward. We also choose in the revised paper to discuss particles larger than 0.01 μm and smaller than 0.01 μm (instead of using 0.005 μm) to avoid any confusion.

RC: Also, choosing delta t=20 minute for weak precipitation in Equation (3) is questionable as it takes several hours (~12 hours) to washout the reasonable amount of submicron particles due to drizzle, otherwise the same amount of aerosols get scavenged by heavy precipitation just in an hour of elapsed time.

AC: The reviewer is correct that it takes hours under weak precipitation to scavenge a reasonable amount of submicron particles. Thus, in field experiments, the time period used to measure particle concentrations (before, during, and after rain events) has to be longer than a few hours in order to generate any meaningful scavenging coefficient values; e.g., the amount scavenged has to be larger than the instrument detection limit and the measurement uncertainties.

This practical concern for field experiments, however, will not be an issue in theoretical model studies. Using a longer or shorter time step will not cause any difference in the calculated scavenging coefficient (from Eq. 3) using modelled concentration if the particle concentrations do not decrease significantly during the time period selected. However, for particles that are scavenged very quickly (e.g., particles larger than 3 μm), using an overly large time step will generate scavenging coefficients that differ from the real value if the same formula (Eq. 3) is used, due to the exponential decay of particle concentration during the precipitation scavenging process.

The major reason for choosing a time interval of 20 min (or less) in calculating scavengign coefficient is because the precipitation intensity often changes significantly during a precipitation period of afew hours. In order to compare our modelled results with measurements conducted under specific precipitation rate conditions, it is easier to use a short time interval so the model precipitation rate can be kept within a small range of prescribed values (0.1, 1.0 and 5.0 mm/hr).

It is also noted that the major applications of theoretical ‘scavenging coefficient’ formulas is in three-dimensional Eulerian air-quality models. In these models, a time step of 20 minute is typical. Thus, using this example time interval to discuss our modelled scavenging coefficient also make sense (note that this is not the time step in our detailed microphysics model, in which a time step of 10 second is used) (see discussion in Wang et al., 2010 on this same topic).

To support the above argument and to address the reviewer’s concern, we have added an additional sensitivity test in the revised paper that shows the percentage of concentration change after 3 and 6 hours of precipitation. Two tables have also been added in the revised paper that explicitly show the percentage change of concentrations for several selected representative particle sizes. A comparison of the calculated scavenging coefficients from the original model runs with those from the longer model runs proves our assumption mentioned above. All these new results have been added in the revised paper.

RC: In the diameter range 0.001 to 100 μm for particles, coarser particles or those beyond 10 μm in diameters deposit quickly to the ground by gravitational settling and diffusion as a collection mechanism is negligible for such large particles (5 to 100 μm). Hence, there is no point in considering particles’ size up to 100 μm in model simulations.

AC: We agree with this comment and we have omitted all discussions of aerosol particles larger than 10 μm. Since it is a one-dimensional model, we can afford to use a large number of size bins and our sensitivity tests show that reducing the upper limit size for particles from 100 μm to 20 μm will not influence model results in any significant way for below-cloud processes.

RC: It is hard to believe that assumed droplets distribution in the diameter range 1 μm to 10 mm, droplets <= 200 μm can reach the ground in the form of rainwater for below-cloud aerosols scavenging (as they easily get evaporated in the transit position itself and are of lesser terminal speeds and can not overcome the vertical air motions). It means droplets in the diameter range 1 to 200 μm evaporates in the atmosphere (sub-cloud layers) due to their negligible terminal speeds.

AC: In this cloud microphysics model, one particle group is for aerosol particles and another group is for liquid droplets which include both cloud droplets and raindrops. Thus, it is necessary to use a size range covering very small cloud droplets. Using such small size droplets does not mean that these small droplets can reach the surface (e.g., concentrations for small size droplets are not significantly different from zero at the surface or at levels near the surface). Discussions on this have been added in the revised paper.

RC: Furthermore, the wake behind the raindrop due to turbulent flow may determine the orders of magnitude of the collection efficiency of raindrops at their rear side, whose relevance either need to be included or discussed in the present study.

AC: We agree that many processes that happen in the real world (such as the one pointed by this reviewer) cannot be modelled properly or observed under controlled conditions. Some of these neglected processes have the potential to influence the scavenging process. As mentioned in the Introduction section and also discussed in detail in Wang et al. (2010), the enhancement of particle scavenging by turbulence can come from two aspects: (1) increased droplet/aerosol collision efficiency due to turbulence and (2) mixing of particles from the subcloud layer into the cloud layer. If the first aspect is dominant, then the current theoretical scavenging coefficient formulations need to be modified to include this effect; and if the second aspect is dominant, then the existing formulations can be used in chemical transport models since vertical diffusion is already include in the mass continuity equation. The major purpose of the present study is to quantify the second effect. In this revised paper, we have added more discussion about other factors that are not studied in the present paper. We admit that the wake effect was not mentioned in this and our previous study and we have added some discussions in the revised paper and recommend for more studies on this topic.

Specific Comments

Abstract

AC: The information of particle and droplet size range is available in the Methodology section. We prefer to present major conclusions instead of model details in the Abstract.

Introduction

AC: Spelling error is corrected.

Methodology

AC: In this cloud microphysics model, one particle group is for aerosol particles and another group is for liquid droplets which include both cloud droplets and raindrops. The same continuity equation applies to both particle groups. This explanation has been added to the revised paper.

A description of the collection mechanisms contributing to source and sink terms for Equation (1) has been added to the revised paper.

In order to produce rain in a 1-D simulation, Zhang et al. (2004) constructed the vertical advection structure in which vertical wind speeds were taken from field measurements. We have added a brief explanation and cited two more references to support our choice in the revised paper.

The rationale for choosing the delta t can be found in our reply to General Comments above.

Detailed discussions on various mechanisms contributing to calculated scavengign coefficient have been presented in Wang et al. (2010). Since the present study is a direct follow up to our previous study (to test a hypothesis made in the previous study), we refer to that paper for more details. Furthermore, we have added many detailed discussion in the revised paper on various factors that could contribute to the calculated scavenging coefficient (see section ‘Factors to consider’ in the revised paper).

Results

AC: The range of the pre-chosen cloud layer has been added to the revised paper.

Since the majority of field-derived scavenging coefficient values were obtained at heights close to surface, the model-derived scavenging coefficient values used for comparison purposes should be values taken from the lowest model layer. However, considering the possible effect of model lower boundary conditions for vertical transport on the results at the lowest model level, all values shown in Figures 1 and 2 are taken from the 2rd model level (16-m mid-layer height). Note that the CARMA model employs a staggered vertical discretization where the algorithm scheme for vertical transport assigns the variable Kz at full levels while the particle number concentration is assigned to half-levels; that is reason the height of the 2rd model level for concentration is 16m. A brief summary of this information has been added to the revised paper.

Summary and Conclusions

AC: See our reply in the General Comments section above. In the revised paper, particles larger than 0.01 μm and smaller than 0.01 μm are discussed separately. We now choose not to use the size 0.005 μm as a threshold to avoid confusion.

We appreciate the lengthy discussion provided by this reviewer. We have incorporated many of these discussions into the revised text.

We agree that many processes that happen in the real world cannot be modelled properly or observed under controlled conditions. Some of these neglected processes have the potential to influence the scavenging process. Nevertheless, the present study identifies one important process, vertical turbulent diffusion, that contributes to observed aerosol scavenging. More importantly, this study quantified the contribution of this process under different vertical diffusion intensity scenarios. We thus continue to believe that this study makes an important contribution to the scavenging literature concerning an ongoing discrepancy between theory and experiment, and given the additional information provided here, we hope that the reviewer now agrees with us.

Interactive comment on Atmos. Chem. Phys. Discuss., 11, 20375, 2011.