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Title: On the discrepancies between theoretical and measured below-cloud particle scavenging coefficients for rain – A numerical study

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General comment:

This paper deals with the relative contribution of vertical turbulent diffusion to below-cloud scavenging for submicron particles. Authors simulated rain production and below-cloud particle scavenging with a one-dimensional cloud microphysics model. They quantified the contribution of turbulent diffusion to overall size-resolved scavenging coefficient that based on aerosols concentration changes due to rainfall. 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 ($0.1 mmh^{-1}$ and $1.0\ mmh^{-1}$). Also, choosing Δt =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 (Garcia Neto, P. J., Garcia, B.A., Fernadez Diaz, J. M. and Rodriguez Brana, M. A.Parametric study of selective removal of atmospheric aerosol by below-cloud scavenging, *Atmospheric Environment,* **28**, 2335 – 2342, 1994). 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.

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. 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.

Although this work seems to me technically sound because of the simulated rain production and below-cloud particle scavenging case study through a one-dimensional model, but becomes less significant for some assumptions in model simulations and lack of discussion about the wake capture of aerosols at the rear side of the raindrops during light and heavy rainfall episodes. There are number of other concerns mentioned below in the specific comments.

Specific comments

Abstract:
It should be clearly mentioned particles and droplets diameter range used for the model simulations.

Introduction:
P20377L20: Check spelling "existing"

Methodology:
In the equation (1), C(r, z, t) denotes particles or droplets concentrations and r represents both the particles and droplet radii, however, the sizes for particles and droplets are mentioned in terms of diameter ranges 0.001 to 100µm and 1µm to 10mm respectively. Also, V_t (r, z) used for both particles and droplet terminal fall velocities. Clarify these notations.
P20379L9: (Apparently, the -------- the scavenging coefficient is hidden------) "included", but with which collection mechanisms? (Clarify and support with the references).
P20379L17: The basis for two peak wind speeds 0.15 and 0.45ms^{-1} (support with the references).
P20380L7: In equation (3), chosen 6 min and 20 min on what basis?
P20380L10-11: Λ(r) derived contain several scavenging mechanisms, but, dominance of one or two relative to other depends upon the particles and droplet size regimes under questions, which need to be mentioned in brief.

Results:
P20380L15: within the pre-chosen cloud layer (mention range).
P20380L23: Clarify the consideration of 16m mid-layer for the calculation of Λ(r), as compare to profiles of aerosol concentration changes in the sub-cloud layers due to rain.

Summary and conclusions:
It is mentioned on P20382L2 in Results section that the contribution of turbulent diffusion was negligible for very small particles (diameter <0.01µm) under moderate to strong precipitation conditions due to already very high Λ(r) associated with Brownian diffusion. On the contrary, on P20383L11-12, "the influence of vertical diffusion was noticeable for particles smaller than 0.005 µm in diameter under weak precipitation but became negligible when precipitation intensity increased to 5mmh^{-1}". Authors should clarify these aspects.

Atmospheric processes such as condensation, nucleation, coagulation and hygroscopic growth of aerosols and poly dispersed distributions of aerosols in the form of various chemical species in atmosphere (Chate et al., 2003, 2004, 2004a), the fractions of which get scavenged, are the factors for aerosols loss and their production terms in the climate dynamic equation which essentially be discussed for their contributions to the size-resolved scavenging coefficients relative to vertical turbulent diffusion [Chate D. M., Rao, P.S.P, Naik M.S., Momin G. A., Safai P.D., and Ali K., Scavenging of aerosols and their chemical species by rain, *Atmospheric Environment,* **37,** 2477 – 2484, 2003; Chate D. M. and T. S. Pranesha, Field studies of scavenging of aerosols by rain events, *Journal of Aerosol Science*, **35**, 695 – 706, 2004; Chate D. M. and T. S. Pranesha, Field measurements of sub-micron aerosol concentrations during cold season in India, *Current Science*, **86**, 1610-1613, 2004a.]. They may be significantly responsible for the discrepancy between theory and observations.

Possibility of wake behind the raindrops as a contributing factor (as wake capture of aerosols) to the discrepancies between observations and model results cannot be ruled out during heavy rain and drizzle. For example, the most serious discrepancy pertains to collection on the backside of the drop. The wake behind the drop and stationary eddies in the wake region may be responsible for collecting the submicron aerosol particles at the rear side of the drop. Berg (1970) discussed in detail the collection mechanism of aerosol particles on the backside of the raindrop. Engelmann (1965) found collection efficiencies above unity or up to 2 and attributed them to the collection on the backside. His experiments were conducted under conditions that very closely resembled to those in the atmosphere, and his results therefore are especially significant to role of wake behind the droplets in size-resolved scavenging coefficient. Engelmann explained the large values of collision efficiency by a combination of wake effect and electrostatic charge effects, but both these effects should favor collection of submicron particles (0.01 to 5 μm) rather than larger particles. Berg (1970) suggested that the electrostatic attraction enhances the collection of submicron particles in the wake region of the drop. [Berg, T. G. Owe, Collection efficiency in washout by rain, Precipitation Scavenging, CONF 700601, U.S. Atomic Energy Commission, pp 169 – 186, 1970; Engelmann, R. J., Rain scavenging of ZnS particles, *Journal of Atmospheric Sciences,* **22**, 719 – 727, 1965.]

The amount collected on the backside may be 10 or even 100 times more than that collected on the front side of the scavenger as reported by Asset and Hutchins (1967). [Asset, G., and Hutchins, T. G., Leeward deposition of particles on cylinders from moving aerosols, *Amer., Ind., Hyg., Ass., J.*, 348 – 353, 1967.]. Thus, the wake behind the raindrop may determine the orders of magnitude of the collection efficiency, whose effect is not included in the present study by the authors. The discrepancy between theoretical and experimental results may be due to such factors, which need to be discussed in this manuscript.

In spite of the apparent simplicity of washout of submicron particles as a model and experimental problem, it contains elements those are difficult to approach completely through theoretically or experimentally. For instance, it is not possible to control accurately the experimental data obtained in field observations, particularly in case of submicron particles during weak and heavy rain events. The discrepancy between the model and observations may be due to various factors affecting the field measurements of aerosol number–size concentrations before and after rain. The uncertainties in the data obtained in the field observations during heavy rain events and drizzle and limitations cited aforementioned in model approach are missing in the discussion part of this paper. The relative dominance of turbulence scavenging in this model simulation should be compared with the other mechanisms (Brownian diffusion, condensation, nucleation, coagulation and hygroscopic growth of aerosols and poly dispersed distributions of aerosols in the form of various chemical species in atmosphere, Electrostatic charge effects on aerosols and droplets (Tinsley et al. 2000; 2001)] and their dominance for various particles and droplet size regimes.

In view of aforementioned concerns, I can not support publication of this manuscript in the ACP in its present form.