Reviewer's report on the manuscript by Jones et al. "Below-cloud scavenging of aerosol by rain: A review of numerical modelling approaches and sensitivity simulations with mineral dust", Atmospheric Chemistry and Physics, Manuscript ID: acp-2022-409

The manuscript presents an examination of exiting parameterizations for below-cloud scavenging (BCS) of aerosols by rain in the context for use in GCMs, particularly pertaining to those with a modal representation of aerosols. Simulations of mineral dust using a GCM (the UK Met Office's Unified Model coupled with UKCA-mode for chemistry and aerosols) were conducted, employing a number of theoretical and empirical formulations of BCS rate. The study aims at addressing several questions: 1) the impact of using an empirical vs. a theoretical formulation for the BCS rate on GCM modelled (mineral dust) aerosols, given the large difference in BCS rates between the two approaches; 2) the importance of the additional physical processes that are often missing in existing BCS parameterizations, such as phoresis and rear-capture processes; 3) the impact of assuming monodispersed aerosols in calculating the BCS rate rather than integrated BCS rates to account for the lognormal distribution of modal aerosols; and 4) the impact of mode merging following BCS. The last two questions are relevant to models using a modal representation of aerosol size distribution only. In addition, the authors also proposed a new parameterization for collection efficiency to account for the rear-capture mechanism. The large uncertainty in the parameterization of BCS of aerosols by hydrometeors remains to be resolved. This study explores the sensitivity of GCM simulated dust aerosols to the different formulations for the BCS rates for size-distributed aerosols. The manuscript is well structured. I do however have some concerns and comments (see below) which I hope that the authors can address before the manuscript can be published.

<u>General comments</u>

The sensitivity results from the GCM simulations of mineral dust can be influenced by how some of the other processes are represented in the model. For example, the choice of treating mineral dust particles as externally mixed insoluble particles throughout their atmospheric lifetime in this study limits the wet removal of dust particles to BCS only. However, through atmospheric processing, dust particles can be coated and become internally mixed with other soluble components. They can participate in cloud process and be subjected to in-cloud scavenging (ICS or rainout). The ICS can be particularly important for accumulation mode aerosols at greater distance downwind from the source regions. If atmospheric aging and ICS were considered for mineral dust in the model, would the sensitivity to BCS be reduced?

How is BCS modelled in the UKCA-mode for other soluble and insoluble modes? Would the same Slinn+ph+rc BCS algorithm be used for those aerosol modes also?

A question on the comparison of modelled and observed (measured) dust deposition fluxes (in Figures 7, 8, and 9): are those total deposition fluxes (model and observation), i.e., including both dry (including sedimentation) and wet deposition fluxes? If so, what is the dry-vs-wet dust deposition fluxes based on the model simulations?

Specific comments

Line 110: KQ4 is not just relevant for BCS. With a modal representation for aerosol size distribution, mode merging will need to be considered for any process that is size dependant. Does the default UKCA-mode not consider model merging for such processes as dry deposition and sedimentation, cloud processing, coagulation, in addition to wet deposition?

Lines 388-389: Is mode merging not performed by default in UKCA-mode (following those processes that affect aerosol size distributions)?

Line 451: Figure 2 is intended to illustrate the relative importance of various physical processes/mechanisms contributing to the overall collection efficiency and BCS rate over the range of particle and rain droplet sizes. However, how do you define dominance here? Is it the one with largest collection efficiency numerically (since it only identifies a single process at any given particle and droplet size)? It would be more instructive to show the contributions from the various processes/mechanisms to the total collection efficiency over the particle size spectrum at some given droplet size (e.g., 0.1 and 1 mm, perhaps). How does the result here compare with the review of Wang et al. (2010) (i.e., their Fig. 1); is the colour label switched over between interception and inertial impaction? Also, the area where the rear-capture mechanism is dominant on Figure 2 seems to be pasted on; the contours of the total collection efficiency seem to be discontinuous there.

Lines 471-472: Note that the 90th percentile fit of Wang et al. (2014) also accounted for the variability from droplet number density and fall velocity formulations.

Lines 491-493: The discussion on aerosol median diameter converging over time is unclear. The authors seem to be referring to the crossover between Λ_N and Λ_M shown on Figure 4.

Lines 505-506: It is curious that Figure 5(b) shows the least change in the accumulation mode diameter after 3-hour integration for Slinn+ph+rc and Slinn+ph+rc(1M), while Figure 5(a) shows the most mass loss for Slinn+ph+rc and Slinn+ph+rc(1M) amongst the non-observation-based BCS schemes. Any explanations?

Lines 590-591: The authors made a comment here about the simulated DOD from LAAKSO being significantly biased low compared with observations, particularly over secondary source regions. The low bias is apparent from Figure 7(f); however, the specific locations of the low bias is not obvious from the said figure.

Lines 592-594: Again the authors are referring to Fig. 7(h) and (i) for the discussion here on where the modelled surface dust concentrations are biased low or high from LAAKSO and WANG compared to observations. However, such information is not indicated from these figures (unlike the scatter plots for dust deposition).

Lines 625-626: Same here, how can you tell the from Fig. 8(g) that the modelled dust concentrations (SLINN in this case) are higher than observations away from source regions?

Lines 658-660: It is not just the wider model width for the coarse mode that are attributable to the greater difference in model results between the double moment vs. single moment approach. The accumulation mode covers the range of particle spectrum where the overall BCS rates are less sensitive to particle size than the size range where the coarse mode covers (ref. to Figure 3).

Line 756: Again, KQ4 is not just relevant to BCS.

Lines 759-761: How is BCS modelled in the default UKCA-mode dust setup?

Lines 775-777: Wang et al. (2014) did not include any rear-capture parameterization in developing their semi-empirical model.

Lines 781-784: The relatively muted effect of the rear-capture mechanism (with regard to the modelled dust metrics) may also be consistent with the relatively narrow range in the droplet sizes when the mechanism is important as shown in Figure 2, as well as possible buffering effects of the multiple processes in the model influencing the overall simulation results.

Lines 785-787: Suggest removing this statement as the argument here is not a reasonable one. The overall collection efficiency is a linear combination of all the collection efficiencies representing each individual physical processes/mechanism in BCS. The inclusion of the phoresis processes would not mask the contribution from the rear-capture mechanism.

Lines 787-788: Table 2 seems to indicate that the significant reduction in modelled global accumulation-model dust burden is mainly due to the addition of the phoresis processes, (rather than a combination of phoresis and rear-capture).

Line 805: What is the BCS scheme used in CLASSIC?

Lines 810-813: How can you tell the model over-predictions of the dust surface concentration in the scatterplot (Figure 11h) are over areas away from the dust source regions?

Line 859-860: Do we know whether the observational derived BCS rates are free of ICS influence?

Minor comments

It would be good to be consistent in referring to $E_{br,in,im,th,df,es,rc}$ as collection efficiencies throughout the text rather than switching between collection and collision efficiencies at different places.

Equation 3a: should it be $N(D_d) = N_0 e^{-\lambda D_d}$?

Line 484: Replace “uniform size distribution” with “monodispersed aerosols”?

Line 485: You mean Eqs 4 and 5 (not 3 and 4)?

Lines 487-488: Could be reworded to “It is clear that the *effective* number (Λ_N) and mass (Λ_M) scavenging coefficients for lognormal aerosol distribution are significantly greater than the scavenging coefficient (Λ) for monodispersed aerosols, …”.

Line 732: Referring to Fig. S12c rather than Fig. S10c?

Line 774: Use “compensating errors” rather than “competing errors”?