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Interactive comment on "A new aerosol wet removal scheme for the Lagrangian particle model FLEXPART" *by* Henrik Grythe et al.

Anonymous Referee #2

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1 general comments

The authors discuss a new parametrization for FLEXPART of both in-cloud and below-cloud wet scavenging of aerosols. The parametrization has revised the way that cloud information is obtained and processed and considers both size dependent, aerosol dependent and phase dependent scavenging. Results from the scheme are compared against observations and results from other published studies for three different aerosols. The sensitivity of the parametrization to the various parameters of the scheme is further assessed.

I found this an interesting paper which is well written and comprehensive.

2 specific comments

1. There is a good, explicit and easy to understand summary of what the new aerosol wet removal scheme includes but I'd like the authors to emphasise further what improvements this new scheme gives over the previous scheme (i.e., which of the features discussed are new), particularly since some discussion of results with the old and new schemes are included.

2. I would disagree with the authors' comments regarding aerosol schemes in Lagrangian models (lines 83-86). Whilst these schemes may have remained constant for some time, developments and reviews have taken place since their first introduction. For example, NAME has recently had a new size dependent wet deposition scheme added for particles.

3. Presumably some critical value of $CTWC$ is used to determine cloud ($CTWC$ > some value). What is this value and how much certainty is there in it? How sensitive are results to this value?

4. Scavenging from multiple layers of cloud may be difficult to accurately model with only surface precipitation data. For example, the precipitation rates / intensities are likely to be different from clouds at different layers. Furthermore, what happens between layers? Is this considered as "below cloud" scavenging given that all layers of cloud are considered to be precipitating?

5. Where does the value of 6.1 for ic_r come from (line 160)? Is it tuned to data? It sounds like results are quite sensitive to the value of ic_r (lines 162-163).

6. The authors might like to mention the Greenfield gap (i.e., that there is a range of particle sizes which are neither efficiently collected by Brownian motion nor impaction) (lines 202-203).

7. The authors use either the surface precipitation type or a surface temperature to determine whether the Laakso or Kyro parametrization should be used. However, precipitation which is rain at the surface may be snow at the point of impaction (or vice versa). Hence it would seem more appropriate to use the temperature at the aerosol height. Also, how is the water phase of the clouds determined (lines 357-361)? Is this determined from the temperature and/or precipitation type? And if so, can the authors comment on the height of data that is used and the appropriateness of this data if surface data is used to determine the phase of elevated clouds?

8. Where do the values for CCN_{eff} and IN_{eff} come from? Can the authors add some references in addition to those given for black carbon? For soluble aerosols Croft et al. (Atmos. Chem. Phys., 10, 1511–1543, 2010) have a large difference between values for liquid and ice stratiform clouds (albeit similar values for convective) which contrasts here to the default values in Table 3 for ^{137}Cs attached to sulfate. I refer the authors to their comment on lines 517-518 questioning whether their in-cloud scavenging in ice clouds is too effective for this aerosol. And lastly, how is it possible for CCN_{eff} (a fraction) to take values > 1 (Table 4)?

3 Technical corrections

1. Line 121 – There is no paper by Stohl et al. from 2016 listed in the references. Is the date here incorrect?

2. An explanation of C^* should be given when equation 4 is introduced.

3. BRW should be BAR (line 321) and Table 4 (or maybe BAR should be BRW on lines 315 and 321).

4. Could the authors label day 7 on Figure 4.

5. A couple of comments regarding Table 2: (i) For 18.2 μm the lifetime for the default parameter settings is given to 2 decimal places, whereas for all other parameter settings, it is only given to 1 decimal place. Furthermore, if the lifetime for the default parameter settings was given to 1 decimal place, it would be the same (0.3) as the lifetimes given for other parameter settings so it is not obvious that there is any change. Is there? Perhaps there should be a consistent use of decimal places here. (ii) The longest lifetime if only one deposition process is reduced is indicated in blue but yet there are many such cases with the same lifetimes. Should these also be indicated in blue?

6. I think the reference to Table 1 on line 421 should be to Table 2.

7. There is an inconsistency in the lifetime increases on line 424. The 20% increase (6.2 μm particles) means that the end result is 120%. The comparison is for a 350% increase (not a 450% increase as stated – this is the end result).

8. The footnote on page 13 states that $ic_r = 6.2$ is used in this paper, whereas the value stated on line 160 is 6.1

9. Is there a typo in the figure caption of Figure 5b? Should “surface area distribution” be “size distribution”?

10. There is a typo in line 473. 10.8 days is for the 0.65 μm bin size (or alternatively 11.7 days for the 0.4 μm bin size).

11. Table 4: The column header refers to “mean” concentration, whereas the caption refers to the “median” concentration.

12. Line 550: “concentration” should be “column burden”.

13. Line 574: Reference to Fig 6 should be Fig 7.

4 Minor issues

Some further minor issues and questions which the authors may also like to address.

1. Lines 4-6 implies that differentiating between cloud water phases allows an aerosol type dependent removal scheme. These seem two independent things.
2. It is not clear whether the reference to "dry deposition" in most places includes "gravitational settling". In the abstract (lines 19-20) the two are referred to separately (Dry deposition and gravitational settling) but gravitational settling isn't always referred to elsewhere and hence one is unclear whether references to dry deposition intend to include gravitational settling.
3. It would help to briefly define "aerosols" as this term is sometimes misused and confused with the generic term "particles".
4. What is the meaning of "outside a cloud" (line 328)? In particular, are not 'below' or 'above' clouds 'outside' of clouds?
5. It seems a little counter-intuitive that there could be more below-cloud impaction scavenging events around 5000 m using the FLEXPART relative humidity parametrization (Figure 3b) when the cloud-top height is, in general, lower using this scheme. The authors mention multiple cloud layers being the reason for the small peak in the below cloud impaction events at this height using the ECMWF parametrization, and that in reality there is probably not scavenging at this level due to the upper level clouds being non-precipitating. Do the same reasons apply to the peak in below-cloud scavenging events at 5000 m seen when using the FLEXPART relative humidity parametrization and why does this scheme give a larger count? The smaller count for the ECMWF parametrization, if the below cloud scavenging events at 5000 m are not real, may be indicative of better performance of the ECMWF parametrization.

6. Is mention of "particles in the 0.2 – 0.6 μm" (line 404) intended to be a general reference to accumulation mode particles or a reference to particle sizes modelled here? If it is the latter, I am having trouble matching this with the numbers in Table 2 which gives a diameter of 0.2 μm but the next size up is 2.2 μm.
7. Nanometre and micrometre are used interchangeably (e.g. 0.2 μm and 200 nm are both used). Some consistency referring to particles of the same size would be better.
8. In the comparison of the measured and model size distribution (lines 447-448), the authors say that the measured peak at 1 μm matched the model well. The aged model peak is at a little lower particle size distribution.
9. What are the upside down black triangles in Figure 5c? Are these the "daily median ratios" (line 455)?
10. The mention of "aerosol type" on line 512 refers, I think, to splitting the aerosol size distribution up into bins with different particle sizes and scavenging rates. If this is correct, I find this terminology confusing since I would say that it is one aerosol type (i.e. ^{137}Cs attached to sulphate) and not more than one aerosol type.
11. Is the dotted black line in Figure 5d the 1-1 line or the line of best fit to the ^{133}Xe (or both?)?
12. I don't follow the final sentence of the paragraph on lines 559-561. I cannot see a burden with a substantially different dependence on latitude in Figure 6. Furthermore, simulations 5 and 8 also have a phase dependent change to the removal parameters.
13. Lines 570-571. The authors refer to an increase in remote areas like the Arctic but the increase seems much larger near the equator.

14. It is difficult to see the black circles in Figure 7, particular since the coastlines are also black.

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