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

Henrik Grythe et al.

zhg@nilu.no

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We would like to thank both reviewers for their detailed and constructive comments on our manuscript. It is very much appreciated that both reviewers took such obvious care and gave excellent comments. It is our hope that they agree that the changes introduced in the new version based on their comments and suggestions, have helped improve the work. Where comments from both reviewers address the same issue, one answer is given for both comments. Below, we list the reviewer comments and our corresponding replies (in blue) as well as excerpts from the new draft (blue italic). A supplement of tracked changes is also provided.

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Discussion paper

RC #1

General comments: This paper describes the incorporation of a new aerosol wet scavenging scheme into the FLEXPART model. The authors included a new parameterization for wet removal within and below clouds considering the physical state of water in the clouds and the precipitation type. This parameterization was set for three different types of aerosols and compared against available measurements. A series of sensitivity analysis were also performed to test the range of results obtained under different parameterization assumptions.

Specific comments:

- Although it is very commendable the inclusion of comparisons between measurements and model results, I have serious concerns regarding the uncertainties in other processes (e.g. emissions) that might hinder the conclusions reached by this work. Consequently, to give the readers a sense of the relative changes introduced by the new parameterizations, I would suggest including the results from the old parameterization for each of the three applications presented in this work.

This is a good and valid concern. Therefore, as suggested we have added some results from version 9 of FLEXPART to each of the 3 sensitivity studies. The following changes were done to accommodate this:

1. In Figure 5c: The ratio of 137CS/133Xe from Kristiansen et al., (2015) using FLEXPART version 9 has been added to show the difference in e-folding timescale from previous versions. In Figure 5d: a fit to the latitudinal bias of FLEXPART v9 is now also shown.
2. For mineral dust, a line was added to Table 2 that reports τ_F using the standard

removal in FLEXPART v9.

3. In Figure 9, we have replaced simulation #7 concentrations obtained with version 9 of FLEXPART. The removal used is however, the removal used for BC in previous publications, which is a modified version of the existing scheme in FLEXPART.

When adding these results, also some small changes to text were made throughout the document to incorporate the results in the text (see supplement).

- Line 160. How did you come up with a value of 6.1 for icr? Is this basically an empirical factor?

The empirical nature of the value of i_{cr} used was perhaps not expressed explicitly enough. Though i_{cr} should be representative of that there is cloud water replenishment, linking the time averaged cloud water fields to precipitation rates, the value of 6.1 is a purely empirical factor in FLEXPART. Values suggested in literature for cloud water replenishment suggest 15-120 min for warm marine stratiform drizzle (Wood et al., 2009). The value of replenishment is closely tied to cloud droplet autoconversion rates (Khairoutdinov and Kogan 2000) which are not as well constrained for mixed and ice phase clouds and not resolved in FLEXPART.

In text it was clarified that i_{cr} indeed is an empirical value:

where F_{nuc}, the nucleation efficiency, is the fraction of the aerosol within the cloud that is in the cloud water (see Fig. 1). While ic_r represents the cloud water replenishment rate, it cannot be determined from the ECMWF output data. Therefore, the determination of the constant ic_r was done on the basis of empirical testing in FLEXPART and must be considered a tuning parameter.

Compared to the previous FLEXPART scheme described in Stohl et al., 2005,

ic_r/PCW replaces the cloud water representation that was calculated based on an empirical relationship with precipitation rate ($cl = 2\,10^{-7}\,I^{0.36}$). The overall best results were obtained for ic_r set to a value of 6.1 for the ECMWF cloud water fields, which is used for all simulations in this paper. This resulted in a somewhat slower in-cloud removal rate with the new compared to the old parametrisation. Comparison of the two parametrisations also shows that using ic_r/PCW gives overall weaker dependence on I, compared to cl in the old removal scheme. For simulations where in-cloud removal constitutes a large fraction of the removal, i.e. especially for soluble accumulation mode aerosols, the empirical value of ic_r has a large impact on overall removal rates.

- Including a list of recommended values for the parameterizations for different aerosols will enhance the value of this work.

This is a good suggestion which we wholeheartedly agree to. A paragraph has been added in “Conclusions” to this effect:

Simulations for the accumulation mode particles with FLEXPART are highly sensitive to the choice of CCN_{eff} and IN_{eff} values, which describe the particles' efficiency to serve as cloud condensation and ice nuclei. Overall, it was found that the sum of $CCN_{eff} + IN_{eff}$ is more important for the removal efficiency than the individual choice of values for CCN_{eff} or IN_{eff}. For the three aerosol types, we recommend the following values: Regarding insoluble aerosols, Zwaaftink et al., 2016 found good agreement between modeled and observed concentrations when using $CCN_{eff} = 0.15$ and $IN_{eff} = 0.02$ for mineral dust. For BC, $CCN_{eff} = 0.9$ and $IN_{eff} = 0.1$ gave the overall best results, and these values are also comparable with what was found by Cozic et al., 2007. Soluble aerosol (^{137}Cs) concentrations compared best with $CCN_{eff} = 0.9$ and $IN_{eff} = 0.9$. The latter value is somewhat higher than IN_{eff} values suggested by measurements of e.g. Henning et al., 2004.

Technical corrections:

Line 40. Please add chemical processes for completeness in the sentence.

Thank you, this is now changed.

Line 85-86. HYSPLIT has a new option for in-cloud wet scavenging parameterization (See Stein et al 2015, supplement). NAME has also updated its wet deposition scheme (see http://www.metoffice.gov.uk/media/pdf/c/a/FRTR584.pdf)

The paragraph has been updated so that it now more correctly reflects that the wet removal of NAME and HYSPLIT has been updated. We now write:

The aerosol removal scheme in FLEXPART Hertel et al., 1995 has remained relatively unchanged since its incorporation in the late 1990s. Other, similar Lagrangian models like NAME and HYSPLIT have had recent updates to their aerosol removal (Webster et al., 2014; Stein et al., 2015). However, the overall level of detail also in these models remains low compared to known theory (e.g. Feng 2007). One reason for this is the limiting factors that constrain the possible ways of treating aerosol removal within the Lagrangian model framework.

Line 91 non-linear chemistry has been included in this kind of models (e.g. Chock, D. P., and S. L. Winkler, 1994: A particle grid air quality modeling approach: 1. The dispersion aspect. J. Geophys. Res ., 99, 1019–1031, doi:10.1029/93JD02795. Chock, D. P., and S. L. Winkler, 1994b: A particle grid air quality modeling approach: 2. Coupling with chemistry. J. Geophys. Res., 99 (D1), 1033–1041, doi:10.1029/93JD02796.)

We have rephrased the paragraph to show that more extensions to the linear model concept exist. We now write:

A main consideration within this framework is that each transported computational particle is independent of others. Extensions of this concept to allow for non-linear chemistry exist (Chock et al., 1994a,b), also for FLEXPART (Cassiani et al., 2013), but the reference version of FLEXPART is a purely linear transport model. Within such a linear model, it is impossible to include aerosol processes which depend on the aerosol concentration (e.g., coagulation or non-linear chemical reactions). Furthermore, to facilitate consistency between forward and backward runs of FLEXPART, parameterizations that depend on the age of the aerosol (i.e. time after emission for primary aerosols) should be avoided as well. This limits the level of sophistication that can be incorporated into an aerosol removal scheme. Nevertheless, a realistic treatment of aerosols is possible even with these limitations.

Line 247. Sulfate is not a primary aerosol. Please correct the sentence.

Yes that is correct, thank you, this is now changed so it no longer reads that sulphate is a primary aerosol.

Line 520- 524 This is very speculative. There is no empirical evidence that this is why the model shows a latitudinal bias.

Agreed. Results presented in the paper do not show any evidence for this, and it is thus a speculation. The paragraph has changed to reflect this. "The probable cause" is changed to "One of the possible causes". Also added a further sentence on other possible causes of the latitudinal dependence of model/measurement bias to highlight that the reason for the latitudinal bias. We have now written:

In Fig. 5d the mean model / observed concentration ratios at the different stations are plotted against latitude. A prominent feature of FLEXPART and indeed most models used by Kristiansen et al., 2015 is a tendency to overpredict concentrations at low latitudes and underpredict concentrations at high latitudes. This tendency is also present *with the new removal scheme, where model / observation ratios decrease with latitude. The green line shows a logarithmic fit to the station median data. The same fit was done to the mean from a simulation using FLEXPART version 9 (pink). This shows that the new model, while still having a systematic latitudinal dependence, represents a clear improvement over the old version. One possible explanation of the decreasing model/observation ratios with latitude might be that in-cloud scavenging in ice clouds is too effective. However, sensitivity simulations where only IN_{eff} was reduced (not shown) revealed that this change had only a small effect in further reducing the latitudinal bias. One of the possible causes of this is the high proportion of mixed phase clouds (77%) which reduces the impact of the latitudinal dependence of the frequency of ice-phase clouds after that much time for an emission pulse. Another possibility is that cloud phase is not well captured by the ECMWF model, as in many other models Cesana et al., 2015. It may also be relevant that the clouds have on average higher cloud tops near the equator, so that temperature and thus the mixing state of clouds does not have a strong enough latitudinal dependence in the Northern Hemisphere at the time of this simulation (March-May).*