Reviewer 1

GENERAL COMMENTS

This manuscript presents a new version of the Lagrangian particle dispersion model Flexpart. The main changes for this new version are described and relevant examples are given for several aspects of the model. The new version is compared with the previous version in terms of computational performance and accuracy by using observations obtained from two tracer experiments. As the Flexpart model is widely used by many researchers for several applications, the manuscript is of high interest to the community. The manuscript is well written and figures and tables are neat. The methods and assumptions are clearly described or properly referenced.

We sincerely thank the reviewer, Pieter de Meutter, for taking the time to thoroughly review our manuscript. We appreciate your detailed feedback and constructive suggestions. We have carefully considered your comments and have made the necessary revisions to address the points you raised.

In the following, responses are in blue, and quoted text is show in green. Text after the little arrow '→' is newly introduced or modified manuscript text in the reaction to the reviewer's comments.

SPECIFIC COMMENTS

l 90: Could the authors add instructions/hints to install Flexpart-11 and its dependencies (or refer to the online documentation)?

We have clarified this by changing the following statement: "Accompanying this paper is a completely revised technical documentation of FLEXPART (https://flexpart.img.univie.ac.at/docs)" → "Accompanying this paper is a completely revised technical documentation, including a download and installation guide, of FLEXPART (https://flexpart.img.univie.ac.at/docs)"

l 95: "FLEXPART calculates particle trajectories using interpolated meteorological fields": the interpolation is performed in time and space?

We clarify this: "FLEXPART calculates particle trajectories using meteorological data interpolated in time and space from time sequences of three-dimensional fields of, e.g., wind velocities, density, temperature, specific humidity, cloud liquid and ice water content."

l 101: "All units are in International System of Units (SI) units, unless otherwise specified.": the context for this sentence is not very clear. Does it relate to the input meteorological data?

We removed this sentence and report units throughout the text.

l 149: "In addition, to avoid regions with low Coriolis force…": could the authors provide a motivation for avoiding regions with low Coriolis force?

We changed the text as follows: → "We only used particles outside the subtropics and tropics, excluding the zone between 40° S and 40° N, as we expect the tracer conservation in this region to be worse in general, where the geostrophic balance is weak and deep convection is frequent."

l 157: "N is the total number of particles in the sample": sample refers here to all particles in the domain between [-80, -40] and [40, 80] latitude?

We rephrased the text as follows: "...where γ_i(t) and γ_i(t_0) are the quasi-conserved property of particle i at time t and t_0 of its trajectory, respectively, and N is the total number of particles in the sample." → "...where γ_i(t) and γ_i(t_0) is the quasi-conservative property of particle i at times t and t_0 of its trajectory, and N is the total number of particles in the domain between [80° S, 40° S] and [40° N, 80° N] latitude that fulfilled our selection criteria."

Figure 1: can the authors provide an explanation why the improvement in the stratosphere is so large for potential temperature compared to the improvement for potential vorticity and specific humidity?

It is true that the improvement for potential temperature in the stratosphere (61 %) is much bigger than that for absolute humidity (17 %) and PV (15 %), and also bigger than the improvement for all three quantities in the troposphere. The answer might lie in the way how errors in the vertical position (which is mainly affected by the new coordinate system) translate into errors of the quantity under consideration. Potential temperature gradients in the stratosphere are much larger than those of absolute humidity, and on top of that, absolute humidity is likely to be more uncertain already in the ECMWF analyses as values are so low. This does not explain why the relative improvement is also low for PV. Here, we have to search the reason in the large absolute values of PV especially in the upper stratosphere.

Figure 2: the authors provide a number of possible causes for deviations from the well-mixed criterion. For |lat|>66° and between the surface up to +-1.5 km, the eta coordinates seem to result in a worse performance in terms of well-mixedness than the z coordinates. Do the authors have any possible explanation for this specific behavior?

Unfortunately, we do not have an explanation. It is generally difficult to trace errors in well-mixedness to a specific reason. They are likely due to several different error sources (interpolation errors, numerical errors, mass consistency of driving ERA5 data) that partly lead to compensating effects. Furthermore, the deviations in well-mixedness may have their origin in completely different regions than where they materialize. The volume of the atmosphere where the z coordinates seem to lead to better well mixedness is quite small (polar regions below some 1.5 km), so we think this should not be overinterpreted.

Figure 3:

- there is only one sentence of discussion for Figure 3, so I suggest to elaborate the discussion a bit or consider omitting the figure.

Discussion of Figure 3 has been extended: '...for releases from Vienna, Austria, spherical particles with a diameter of 50 µm are deposited mainly in Central Europe, whereas volume equivalent fibres with an aspect ratio of 50 are also deposited in Eastern and Southern Europe (Figure 3a,b). The longer atmospheric transport of fibres results in an average horizontal travel distance 2.7 times greater than that of spherical particles (Figure 3c).'

- the results shown in Figure 3 represent total deposition, that is, the combined effect of dry and wet deposition. Wet deposition will diminish the relative difference between the total deposition of spheres versus fibers? Therefore, it would be useful for the reader to have an idea of the amount of precipitation in January 2018 (that is, was it particularly dry or wet in that period?).

In the case of the release shown in the 'Gravitational settling' section, more than 90% of the total deposition is dry deposition, which is shape-dependent. This is now mentioned in the caption of Figure 3: "In both cases, dry deposition accounts for more than 90% of the total deposition". Note that this specific behaviour is due to the large size of the released particles and their large settling velocities. For smaller particles (e.g., accumulation mode particles), wet deposition would dominate the total deposition.

l 251: "Experiments show that non-spherical particles experience a larger drag in the atmosphere and therefore have lower settling velocities than spheres (Tatsii et al., 2024).": this is assuming identical particle volume and particle mass?

Yes, identical particle volume and particle mass are assumed. The sentence has been changed accordingly: 'Experiments show that non-spherical particles experience a larger drag in the atmosphere and therefore have lower settling velocities than volume-equivalent spherical ones (Tatsii et al., 2024).'

l 273: "PLA=2 PIA=2 PSA": do the authors mean "PLA=2, PIA=2 * PSA"?

No, the sentence has been changed for clarification: '...characterized by PLA = 2 · PIA = 2 · PSA.'

Figure 4: Comparison of panel (a) and (c) seems to suggest that in panel (c) wet deposition occurs even if there would be no rain in the meteorological data. Could the authors comment on this?

Gaps between areas with deposition in (a) are mainly caused by the lack of interpolation for wet-deposition parameters, chiefly precipitation fields, in v10. The fact that the movement of *precipitation systems can be fast enough to skip grid cells within the 3 h interval between fields contributes as well. Therefore, it is (a) which is wrong, not (c). In addition, it is obvious that the pattern in (a) is unphysical whereas that in (c) looks realistic.*

Subsection 5.2: it is not clear to me whether the underlying problem with the interpolation relates to time, space or both. In the text, the focus seems to be on the temporal interpolation, while I thought the problem was more related to the fact that precipitation represents a grid box average value rather than a point value?

The problem exists both in time and space. However, the problem in time is the bigger one, because

1) the central time for precipitation and the time for all other met fields do not match, and the workaround used up to now smoothes the precipitation time series, leading to a spread of precipitation into precipitation-free intervals (on top of what interpolation may do), as explained in Hittmeir et al. 2018.

2) Comparing spatio-temporal structures of precipitation fields and the actual spatial and temporal resolution of the met data shows that the difficulty to properly represent reality is greater in the temporal dimension.

We consider it desirable to extend the approach also to space, as briefly outlined in Hittmeir et al. 2018. However, this is not yet ready for implementation.

In addition, could the authors briefly mention the two meteorological fields (large scale precipitation and convective precipitation?) that are used when numpf = 3?

Large scale and convective precipitation are always read in. The two additional fields are rather additional in time and this refers to both LSP and CP fields. We clarified this in the following way: "This causes FLEXPART to read in two sets of precipitation fields (a set of three large scale and convective precipitation fields each, making it six fields in total) per input time interval instead of one. These additional fields represent two additional supporting time steps in between the original ones to represent precipitation as point values and, at the same time, preserve the integral precipitation in each time interval, guarantee continuity at interval boundaries, and maintain non-negativity (see Hittmeir et al., 2018, for more details)."

Subsection 5.3: the fix for the presence of clouds in case of convective precipitation seems quite arbitrary? Could the authors give an idea of the sensitivity of the choices and whether they think this is a large source of model uncertainty or not? Lastly, is there a particular reason for starting the cloud at ground for convective precipitation above 0.1 mm/h?

It is not totally arbitrary, it is based on some preliminary statistical investigations and meteorological experience.

Sensitivity will very much depend on the scenario, especially the height where particles are mainly located. As said, it is a preliminary, rough fix. Right now, we want to avoid missing wet deposition completely or to a large extent if there are no or only shallow resolved-scale clouds, and still do a little better than with a bulk formula as used up to v6.

We tested the following criteria for a global tropical release of 1 kg consisting of 1 million particles between 0 and 10 km spread between 1 January and 1 February 2017 using ERA5 data:

1. *convp_precmin = 0.1 mm/h, low: 0-10km, high: 0.5-8km. The fix gets called in total 15% of all convective precipitation cases, of which 83% are classified as low and 17% as high, the total deposition is 0.8648 kg, a maximum value of 0.0371E-12 kg/m^2, and a mean of 0.0010952E-12 kg/m^2. (fig: orig.ps)*
2. *convp_precmin = 0.1 mm/h, low 0.1-10km, high: 1-8km. The total deposition is 0.8639E kg, a maximum value of 0.0364E-12 kg/m^2, and a mean of 0.0010941E-12 kg/m^2. (fig: l100h1000.ps)*
3. *convp_precmin = 1 mm/h, low 0.1-10km, high: 1-8km. As expected, the fix gets called again in total 15% of all convective precipitation cases, of which now 99.8% are low and 0.2% high, the total deposition is 0.8624 kg, a maximum value of 0.0357E-12 kg/m^2, and a mean of 0.00109225E-12 kg/m^2. (fig: l100h1000_1mm.ps)*
4. *convp_precmin = 5 mm/h, low 0.1-10km, high: 1-8km. The fix gets called again in total 15% of all convective precipitation cases, of which 99.9997% are low and 0.0003% high, the total deposition is 0.8623 kg, a maximum value of 0.0351E-12 kg/m^2, and a mean of 0.0010942E-12 kg/m^2. (fig: l100h1000_5mm.ps)*

Attached are maps of the four cases, which show no visible differences. We therefore conclude that this is certainly something that needs to be further tested, especially on regional scale, but deposition seems to be not very sensitive to these criteria on a large scale. We added the following warning to the text:

"The results of preliminary tests indicate that the deposition resulting from convective clouds is not significantly influenced by the parameters used in this fix. However, further investigation is required to ascertain the full extent of the influence of these parameters and possible further optimisation."

Figure 5: what is "sum" in the figure?

"Sum" refers to the ratio of the sum of all cs137 and xe133 as defined by eq. 3 in Kristiansen et al. (2012). This gives a more robust measure of aerosol removal times than values taken from

individual stations (see Kristiansen et al., 2012). We added the following clarification to the caption: "Red boxes show the ratio of the sum of cesium-137 and the sum of xenon-133 over all stations."

Subsection 5.4: could the authors provide motivation for aiming for an aerosol decay time of 9.3 d, close to the model median, rather than that based on the observations, which was 14.3 d?

We did not aim for a specific aerosol lifetime, although we agree that the measurements would rather suggest a longer lifetime than the model. However, we do not want to tune the model to only this specific Fukushima case, since we do not know how representative it is for other weather situations or for other aerosol types. Most models have even shorter aerosol lifetimes than FLEXPART.

l 368: "The mixing ratios for each species are calculated using the ratio of the mass of the species over the mass of air, where the mass of air is always carried by species number 1.": does this mean that the mass of air attributed to a particle changes during the simulation? If so, what processes in Flexpart modify the mass of air attributed to a particle?

The mass of air represented by each particle is constant throughout the simulation. Only the mass of other species can vary (e.g. due to chemical reactions, radioactive decay, and fluxes at the surface). This is a special feature of the LCM mode, which is important there because particle densities are typically low in the domain-filling mode, which would lead to noisy results if mixing ratios were calculated by summing particle mass and dividing by air density and volume. Furthermore, LCM simulation times are often very long, and this method avoids problems with the slight violations of the well-mixedness condition in FLEXPART.

l 713: "There also exists a FLEXPART version for very-high-resolution simulations (1 km), where turbulence parameterisations have been adapted to account for the fact that turbulence is partly already resolved by the meteorological input data (Katharopoulos et al., 2022).": this seems quite similar to Flexpart-AROME?

There exist several FLEXPART versions for higher-resolution regional models (e.g., FLEXPART-AROME, FLEXPART-WRF, FLEXPART-ICON, etc.). However, to our knowledge only the Katharopoulos et al. (2022) paper describes a method to explicitly separate between resolved vs. unresolved turbulence.

TECHNICAL CORRECTIONS

l 110: "The ECMWF's IFS employs a hybrid pressure-base vertical coordinates": omit "a"

Corrected

l 193: "byCassiani": add space

Corrected

Figure 3: in the caption: “top” should be “bottom”

Corrected

Figure 4: title in panel (c): consider writing numpf3 rather than npf3 to make it consistent with the text.

We have made the title of panel (c) consistent with the text by replacing npf3 with numpf3.

Table 1: “Number gives the total…”: should be “n gives the total”.

Corrected

l 413: “FA5” should be “FMS”.

Removed sentence in agreement with comments from the other reviewers

Figure 8: the figure size should be smaller.

It should have the width of a column in the published version. We will make it smaller for the next discussion upload.

Table 2: In the caption, move the sentences “Weak scaling is defined by…” and “The serial comparison is done by …” to the text.

Corrected

l 511: omit “solid black lines” (should be in the figure caption or legend).

Corrected

l 512: omit “dashed black lines” (should be in the figure caption or legend).

Corrected

l 581: I suggest to omit “will”.

Corrected

Figure 9: labels (a), (b) and (c) are missing.

Corrected

l 641: Should be “Appendix A”.

It should be Appendix B instead of Appendix (section+B), we have corrected this mistake. Thank you for pointing this out.

l 793: omit the second “of”.

Corrected

l 858: omit “a of”.

Corrected

References:

- Hittmeir, S., Philipp, A., and Seibert, P.: A conservative reconstruction scheme for the interpolation of extensive quantities in the Lagrangian particle dispersion model FLEXPART, Geoscientific Model Development, 11, 2503–2523, https://doi.org/10.5194/gmd-11-2503-2018, 2018.
- Katharopoulos, I., Brunner, D., Emmenegger, L., Leuenberger, M., and Henne, S.: Lagrangian particle dispersion models in the Grey Zone of turbulence: Adaptations to FLEXPART-COSMO for simulations at 1 km grid resolution, Boundary-Layer Meteorology, 185, 129–160, https://doi.org/10.1007/s10546-022-00728-3, 2022.
- Kristiansen, N. I., Stohl, A., Olivié, D. J. L., Croft, B., Søvde, O. A., Klein, H., Christoudias, T., Kunkel, D., Leadbetter, S. J., Lee, Y. H., Zhang, K., Tsigaridis, K., Bergman, T., Evangeliou, N., Wang, H., Ma, P.-L., Easter, R. C., Rasch, P. J., Liu, X., Pitari, G., Di Genova, G., Zhao, S. Y., Balkanski, Y., Bauer, S. E., Faluvegi, G. S., Kokkola, H., Martin, R. V., Pierce, J. R., Schulz, M., Shindell, D., Tost, H., and Zhang, H.: Evaluation of observed and modelled aerosol lifetimes using radioactive tracers of opportunity and an ensemble of 19 global models, Atmospheric Chemistry and Physics, 16, 3525–3561, https://doi.org/10.5194/acp-16-3525-2016, publisher: Copernicus GmbH, 2016.
- Tatsii, D., Bucci, S., Bhowmick, T., Guettler, J., Bakels, L., Bagheri, G., and Stohl, A.: Shape Matters: Long-Range Transport of Microplastic Fibers in the Atmosphere, Environmental Science & Technology, 58, 671–682, https://doi.org/10.1021/acs.est.3c08209, pMID: 38150408, 2024.