
1 Answer to Anonymous referee #2, received 20th may 2020

We wish to thank the referee for his/her helpful comments. The comments of the referee are in bold, our answers in normal black and the changes that have been brought to the manuscript are in blue.

1.1 General comments

This manuscript presents new numerical modeling approaches to represent vertical transport of pollutants plumes in the upper troposphere with Eulerian Chemistry Transport Models (CTM). The aim is to limit the excessive vertical diffusion of the plumes of pollutant in this kind of numerical representation. Different numerical strategies are considered to address this issue: different vertical wind diagnosis, different advection scheme and different vertical resolution. The sensitivity of the simulation of a plume transport event to the different numerical choices considered in this work is evaluated on the case of the Mount Etna’s eruption of March 18, 2012. It is a topic of scientific interest and certainly within the scope of Geoscientific Model Development. The general presentation of the work is logically and clearly organized. However, the added value of this work could be improved with the clarification and/or the development of some results.

1.2 Specific Comments

Section 2.1 p4 l7 and l13: Could the authors precise which CHIMERE version has been used? 2013, 2017 or 2016?

Version has been added in the title: CHIMERE model (v2017r4; Menut et al., 2013; Mailler et al., 2017)

p4 l14: The horizontal resolution of the WRF simulation should be mentioned.

This precision has been brought to section 2.1: The horizontal grid is the same as the CHIMERE grid, with a 5 km resolution.

p4 l17: The authors should provide the limits of the vertical layers (at least in supplement with figure S4).

We agree that some precision was lacking of the model vertical coordinate. However, it would be extremely tedious to provide all the vertical levels, and
these are not directly human-readable since this a hybrid sigma-pressure coordinate. We have added a brief description of the vertical discretization and refer the reader to the publication where the detail of the discretization strategy is provided:

The discretisation of the vertical levels is as described in [?], with vertical levels of exponentially increasing thickness from surface to 850 hPa, and evenly spaced (in pressure coordinates) from 850 hPa. The vertical coordinate depends on the ground-level pressure, with finer vertical levels over elevated ground. The reader is referred to [?] (Section 3.1) for the detailed description of the vertical discretization of the CHIMERE model.

p4 116-17 : There is an in-depth discussion on the relationship between horizontal resolution and vertical resolution in the article by Zhuang et al. (2018) that the authors cite in the introduction, but nothing is said here on this subject.

A discussion of this aspect is already proposed in the introduction, though not bringing it to the same level as in Zhuang et al. 2018:

p3l8 : Apart from this wind-mass inconsistency issue, and more specifically for the representation of polluted plumes that are transported over a long range, Zhuang et al., (2018) have shown that correct representation of long-range transport of polluted plumes in the free troposphere is severely limited by the insufficient vertical resolution. They show, through dimensional and theoretical arguments, that if $\Delta x$ is not at least several hundred times $\Delta z$, representation of long-range transport of plumes in the free troposphere is hindered primarily by this coarse vertical resolution, and increasing horizontal resolution does not bring substantial added value in terms of reducing numerical diffusion of the plume. Since the $\Delta x/\Delta z$ in typical chemistry-transport models is around or below 20 (with a horizontal resolution of, e.g., 20 km for continental scale studies and vertical resolution of, e.g., 1 km), these authors claim that no major improvement will be reached in the representation of long-range transport plumes unless vertical resolution is refined drastically compared to current typical configurations.

In the revised version, the reader is explicitly redirected to that study for a more in-depth discussion of this matter:

For a more detailed discussion of the theoretical ground of this relationship between horizontal and vertical discussion, the reader is referred to Zhuang et al., 2018.

Beyond this point, it is weird to see that the chosen vertical extension of the domain does not provide any possibility to reproduce the highest part of the plume as seen by the observations (cf. figure 2e, S3 and S4). All the more so when we see that the meteorological simulation would allow the domain to be extended. Could the authors explain how they chose the different vertical resolutions tested?

The point made by the Referee is a good point. Our choices were to guided by the idea of choosing typical configurations for chemistry-transport models, including their drawbacks. The Chimere model is not equipped with stratospheric chemistry, and therefore 150hPa is the highest model top value that
can be chosen in the model for realistic simulations. Here of course model top could have been extended further up for the need of this particular study since we use inert chemistry, but we have the feeling that leaving the model top at 150hPa permits us to expose more of the problems that occur in typical use of chemistry-transport models, including the discussion of leakage through the model top. This matter is relevant for the simulation of long-range advection in such models, avoiding leakage of the plume through model top, but also for operational air quality forecast since, as has been shown by Emery et al., (2011), input of stratospheric ozone into the model through spurious mass fluxes at model top significantly affects operational forecast, as discussed in the introduction.

This limitation is explicitly discussed in the revised version:

Section 2.1, The top of model is placed at 150 hPa, with either 20, 50 or 99 vertical layers to evaluate the impact of vertical resolution on the volcanic plume. Even though a higher model top would have been useful for the study of this plume, 150 hPa is a typical value of top of model for CTMs that do not include stratospheric chemistry as it is the case of the CHIMERE model. Also, this relatively low value for top of model permits to examine the question of spurious mass fluxes through the top of model which, as found by Emery et al., (2011) is of relevance not only for long-range transport but also for ozone forecast to ground level.

p4 l17: These different vertical resolutions rely on an oversampling of the same simulated meteorological fields. Could we expect to get significantly different vertical profile with a meteorological simulation carried out with a finer vertical grid?

Our feeling is that the scale of the vertical wind gradients in the free troposphere (a few thousand meters) is larger than the scale of the change in tracer concentration in a volcanic plume (a few hundred meters because, as discussed in, e.g., Zhang et al. 2017, Eastham et al. 2018, these plumes are maintained extremely thin due to the persistent effect of wind shear). However, we are not able to bring forward a proof of this qualitative argument, and to our knowledge a systematic evaluation of the impact of the vertical resolution of the meteorological simulation on plume advection in chemistry-transport models is yet to be done.

p4 l17: Only part of the 33 vertical levels of the meteorological grid is used for the interpolation on the dispersion grid. The number of levels concerned could be specified in this paragraph.

WRF vertical grid has been added to Figure S4 so that the reader can visualize by himself the WRF vertical discretization at the side of the CHIMERE discretization. Also, in Section 2.1, we precise that (28 levels are into 1013-150 hPa range).

p10 l11 and p12 l5: The comparison between the different vertical resolutions involve an aspect which may deserve a bit more detailed discussion. Which kind of boundary conditions are applied for the pollutant concentrations?

We do not have boundary condition for volcanic SO2. This clarification has
been added in the revised version (Section 2.1):

No boundary conditions were used for SO\textsubscript{2} in our simulations.

We feel that this choice is justified because we are interested in the volcanic plume only. We do not simulate the background SO\textsubscript{2} levels. If we would have made the choice to simulate these background levels, then not only an appropriate boundary condition would have been needed but also a proper set of anthropogenic emissions, which was not the purpose of the present study.

With a plume injection in the last layer of the model (at least when 20 levels are used) it seems that the boundary conditions could play a role. What happens when the flux is downward oriented? (here again the choice of a larger vertical extension would be more relevant).

Because there are no boundary conditions used (or, equivalently, the influx of air into the simulation domain has no SO\textsubscript{2} content), mass that is lost through upper boundary can not be brought back into it if wind turns downward. This is an issue in the 20 vertical levels cases, to a lesser extent to 50 vertical levels cases, compared to 99 vertical resolution cases, where little mass is lost through model upper boundary (q.v. Figure 3).

p13 l1-4: Could the authors provide the levels concerned in these tests?

The tests have been done one the 3 vertical resolutions. It had no impact on the 20 vertical levels resolution emissions, as eruption profile width was thinner than CHIMERE top level. Only in the 99 vertical level case was observed a slight difference but not really significant on plume trajectory. It has been specified in the document: p13 l14 The tests have been conducted on 20, 50 and 99 vertical levels resolution.

p13 l5-7: The authors mention an ”injection to a unique altitude”.
It implies the different simulations with the different vertical resolution do not start with the same vertical extension of the plume. It would be interesting to isolate the impact of this initial discrepancy that cannot be associated to an excessive diffusion of the advection scheme. I guess this could be done with a 50 or 99 levels simulations run with an injection uniformly distributed over the different layers corresponding to the injection layer of the 20 levels simulation.

It is possible to see the initial vertical extension of the plume on Figure 7, and indeed, simulations do start with different vertical extension of the plume.

We agree that the point brought to our attention by the Reviewer was deserving additional work. We have performed new simulations with a similar injection profile in all simulations, as the reviewer suggests, and we provide the results of these simulations in Figure S5 of the revised manuscript. This new set of simulations permits to have a better quantitative feeling of the results since avoiding the unnatural offset between the different volume curves. We are particularly grateful to the Reviewer for this suggestion.

A paragraph has been added in the manuscript to describe the results:

To evaluate the impact schemes and vertical resolution would have with a similar vertical extension at injection, new simulations have been conducted imposing an identical vertical distribution at the first time (spreading vertically the emitted mass over the same thickness in the 50 and 99-level simulations than it has in the 20-level simulation). Simulations have been conducted for 20, 50 and 99 vertical levels, for WRFW-DL and NODIV-VL parameters, a total of six simulations. Results have been displayed in supplements, on Figure S5. It can be seen on Figure S5 (left) that all plumes have the same initial volume regardless of vertical resolution, which was not the case in the previous case (c.f. Figure 8a). With a larger vertical extension of the plume at injection,
volumes are higher than in the "unique cell injection" cases, but resolution and transport scheme influence in the same way the evolution of plume (considering its volume). Figure S5 (right) shows evolutions of SO$_2$ highest column vertical profile, similar to Figure 7. This new set of experiments show that, even when getting rid of the initial distortion due to sharper injection profiles in the simulations with the most refined vertical distributions, the increase in plume volume is much slower in the 99-level simulations than in the 20-level simulations. The final volume is about 4 times smaller in the 99-level simulations compared to their 20-level counterparts. A similar factor in volume reduction is obtained by changing strategy from VL-NODIV to DL-REALW. In total, final plume volume in the worst-case NODIV-VL-20 simulation is about 20 times bigger than final plume volume in the best-case WRFW-DL-99 simulation.

**p14 section 3.3** : With the location of "the model column with the strongest vertically integrated SO2 content" the authors have chosen a very aggregated indicator for the comparison between satellite soundings and model results. I assume this choice was made for sake of simplicity in the presentation of the results. However, seeing that the configuration option can lead to some plume splitting, it would be interesting to have more information concerning the horizontal extension of the plume in the different cases.

The aim of section 3.6 *Parameters impact on SO$_2$ dispersion* is to evaluate plume diffusion over 3 dimensions (minimum volume containing 50% of the SO$_2$ mass), and volume results are applicable to surface (cf. p19 l7: *By extension, it has been observed that volcanic plume shape has been modified by DL and WRFW parameters, reducing the surface area containing 50% of SO$_2$ total mass*).

To illustrate the differences, Figure S6 (in suppl.) has been added to show the horizontal dispersion of plume on various simulations after 2 days.

Also, information on the horizontal area of the plume has been added (Fig. S7) and commented briefly in the manuscript: We have also calculated the minimum area containing more than 50% of the SO$_2$ mass (Fig. S7), showing that the WRFW-DL simulations concentrate 50% of the plume mass in an area at least twice as small as their NODIV-VL counterparts.

**p14 l23** : It is not clear to me if the results in figure 5 present average over different configuration options. For instance in the first panel, the simulations with the different advection scheme are compared. Do the number are averages over the different vertical resolutions (the vertical resolution is not mentioned either in the text or in the label of the figure)? Does this imply that there is few interaction between the tested options?

To produce this figure, differences (in km) between model and satellite plumes centroids are calculated for each simulations, then parameters impact are evaluated by calculating the mean between simulation-satellite differences. For instance, "NODIV-DL" (1st line, left column) is the mean between "NODIV-DL-20", "NODIV-DL-50", "NODIV-DL-99". "NODIV-99" (3rd line, left column) is the mean between "NODIV-DL-99" and "NODIV-VL-99". This method has been used to better evaluate the impact of each parameter independently,
Figure 7. Evolution of SO₂ vertical profile (in ppb) corresponding to the maximum column for each step after the Etna eruption, for each tested model configurations. 1st row: NODIV-VL; 2nd row: NODIV-DL; 3rd row: WRF-VL; 4th row: WRF-DL. Left: 20 vertical levels; Center: 50 vertical levels; Right: 99 vertical levels. WRFW simulations values have been corrected to fit NODIV strategy masses.
Figure S5. Left) Minimum volume evolution calculated for 50% of SO₂ total mass in the atmosphere. Right) Evolution of SO₂ vertical profile (in ppb) corresponding to the maximum column for each step after the Elma eruption, for each tested model configurations. 1st row: NODIV-VL, 2nd row: WRF-VL. Left: 20 vertical levels; Center: 50 vertical levels; Right: 99 vertical levels. WRF simulations values have been corrected to fit NODIV strategy masses.

Figure S6. Volcanic plume integrated column dispersion on march 20th at 11 A.M. UTC (2 days after the eruption).
instead of each simulation. The caption has been expended to help the reader better understand the figure:

To produce this figure, differences (in km) between model and satellite plumes centroids are calculated for each simulation, then parameters impact are evaluated by calculating the mean between simulation-satellite differences. For instance, "NODIV-DL" (1st line, left column) is the mean between "NODIV-DL-20", "NODIV-DL-50", "NODIV-DL-99". "NODIV-99" (3rd line, left column) is the mean between "NODIV-DL-99" and "NODIV-VL-99".

From figure 4 we can see that the WRFW-DL-99 simulation is not the closest to the observation at the final stage. This may not be the intuition get from the results presented.

We present the trajectories to explain what the more synthetic results in, e.g. Fig. 5, mean. It is almost impossible for us to visually extract the information from this set of 12 curves and sort out the effect of all three variable parameters in the simulations, this is why we chose to build more synthetic indices and average simulation ensembles together to isolate as much as possible the effect of the parameters without having too much influence of the good or bad luck that can impact every separate simulation.

On Fig. 4, "WRFW-DL-99" is among a set of, say, 4 simulations that are the closest to the observed satellite plume at final stage, but not the closest.

Could the authors precise how the distribution are built? It is not clear for me if it represents different time steps, different locations, or a mix? Are the observations uncertainties are represented in this figure?

Brackets correspond to distribution’s 10th and 90th percentiles (precision now brought to Figures’s caption) and observation uncertainties are not represented in this figure: the figure represents only the spread in the satellite-retrieved altitudes and in the modelled altitudes, for the easter plume (above) and the western plume (below), c.f. Figure 2 and Table 3, column 6th (λthr,i).

These lines are frustrating from my point of view. The authors focus their work on the excessive vertical diffusion in the dispersion model and the only comparison of the model results to observation concerning the plume vertical extension indicates that this plume property is underestimated. Could the authors provide a more in-depth discussion concerning this point? Some considerations concerning the time evolution of the maximum concentration (modeled and observed) could be useful here to convince the readers that a less diffusive treatment of the advection is really suitable. Since the transport in this application is linear, even a normalized comparison to the ”initial” (sounding number 1) maximum concentration would be useful.

We understand the frustration of the Referee about this point, it is our frustration too. However, the large ”brackets” in the 10th to 90th percentiles for the satellite measurements are, unfortunately, due only to large uncertainties in the satellite retrievals. In the same line, estimates of maximal concentration in the satellite data are very uncertain, in part but not only due to the uncertainty
on the vertical profiles of the satellites. This is why we are able to use comparison to satellite data only to check the general structure of the modelled plume but unfortunately not to give a comparison point on vertical diffusion. Even though we are not able to prove it separately in the present study due to inadequacy of our satellite data for this purpose, we consider that excessive vertical diffusion in Eulerian CTMs are already a well-known and very general problem (e.g. Colette et al. 2010, Emery et al. 2011, Zhuang et al. 2018 etc.).

In the revised version, we add a sentence to explicitly state the limits of the comparison to satellite data in link with uncertainties of the latter

The dataset also provides error-range estimates along with the retrieved plume altitude. These error-range estimates have a median of around 1000 m in the western plume and 5000 m in the eastern plume, which is much higher aloft. These uncertainties help to understand the wide distribution obtained from satellite. It is also worth noting that this dataset provides plume altitude but does not provide an information on plume thickness. Therefore, comparison between the left and right panels in Figure ?? does not represent the compared plume thickness between model and observation, but the compared variability of plume height. Unfortunately, due to the relatively large uncertainties affecting the retrieved altitudes, no conclusion can be made on this point either. With all these imitations, Fig. ?? prove that model simulations represent the general structure of the plume, with an elevated eastern plume and a low western plume, and that the median altitudes of both these plumes are very comparable to the median of the satellite-provided altitudes.
1.3 Technical corrections

p5 l11: It seems an "overbar" is missing for notation consistency. It has been modified, thanks

p11 label table 3: The last sentence should probably be in the label of Figure 2.
It has been modified.

p18 l21: erroneous citation