

Answers to the reviewer comments for the paper: "Volcanic sulfur dioxide total columns and layer height from the IASI and TROPOMI Layer height products: preprocessing and assimilation on a 0.1° regional domain of MOCAGE"

We would like to thanks the reviewer for their detailed comments and suggestions, which will help improve the article. In the remainder of this document, we provide our full response (in blue) to the reviewers' comments (in black), which details what we did to address their concerns. We include in this document (in italics) some original passages of the article (in green), with modifications and improvements (in red). The following nomenclature has been adopted for the figures and tables: Fig.X are the figures in the initial version of the article, FigRev.x/TabRev.x are the figures/tables in the new version of the article and FigRep.x the plots used exclusively in this document.

The paper present an evaluation of different approaches to assimilating volcanic observations into the MOCAGE offline chemical transport model. The goal is to improve the simulation of volcanic SO₂ amount and altitude to help in situational awareness (prediction) during volcanic events. Two case studies are evaluated, and several assimilation approaches are tried: simple TROPOMI SO₂ amount, TROPOMI SO₂ and layer height, IASI SO₂ and layer height, and a joint approach that combines TROPOMI SO₂/layer height with IASI. Preprocessing steps are described that are meant to homogenize the observations for suitability in assimilation into MOCAGE. Results are compared (somewhat qualitatively) with OMPS and MSG satellite observations and AirBase surface observations. Including layer height information in the assimilation generally resulted in higher simulated SO₂ abundances than SO₂ amount alone, which is a positive outcome and showed ability to simulate observed surface concentrations for low altitude plume events.

The paper is generally well-structured and well-written. Most of my comments are for improving clarity of presentation, but I do raise a couple of issues that I think warrant further revision. First, the preprocessing approaches described do not seem clearly justified and seem to result in—for lack of a better word—invention of observations that I don't find entirely credible. More discussion on the preprocessing is therefore warranted.

Second, MOCAGE itself seems to struggle with data retention (i.e., SO₂ added is removed very quickly) which is evident especially in Figure 10, which is still somewhat at odds with the high SO₂ values returned. It seems the authors think that plume thickness assumptions are partly the culprit, but I think some further information about the model chemical mechanism is needed (i.e., is SO₂ rapidly scavenged or otherwise converted to sulfate?).

Finally, I think that this paper needs to also present the analysis increments for the various simulations (graphically and numerically) to show where and how SO₂ increments are applied.

These three general comments are addressed in detail in our responses to the comments on lines 294 and 310, line 422, and line 430, respectively. The increments is numerically given in TabRep1. for the case of the Mount etna eruption.

Line 56: Was not familiar with AirBase. Please just briefly describe here what that is. We add a sentence to describe this network.

The AirBase network is a European network of ground-based air quality monitoring stations. The AirBase network contains a large number of stations and covers Europe. These stations measure

Date	ref			tropomi_lh_assim			iasi_assim			joint_assim		
	> 0	< 0	Sum	> 0	< 0	Sum	> 0	< 0	Sum	> 0	< 0	Sum
2024080409	-	-	-	-	-	-	1.000	0.000	1.000	1.000	0.000	1.000
2024080410	0.007	0.000	0.007	-	-	-	0.334	-0.152	0.182	0.334	-0.152	0.182
2024080412	0.508	-0.006	0.502	1.598	0.000	1.598	-	-	-	1.151	0.000	1.151
2024080419	-	-	-	-	-	-	0.690	-0.121	0.569	0.499	-0.264	0.236
2024080420	-	-	-	-	-	-	0.433	-0.174	0.259	0.381	-0.226	0.155
2024080508	-	-	-	-	-	-	0.788	-0.044	0.744	0.737	-0.070	0.667
2024080509	-	-	-	-	-	-	0.116	-0.037	0.079	0.097	-0.047	0.050
2024080510	0.207	0.000	0.207	-	-	-	-	-	-	-	-	-
2024080512	0.540	0.000	0.540	-	-	-	-	-	-	-	-	-
2024080518	-	-	-	-	-	-	0.302	-0.007	0.295	0.301	-0.008	0.293
2024080519	-	-	-	-	-	-	0.254	-0.061	0.194	0.257	-0.061	0.196

TabRep1.: SO₂ increments (analysis-background) for the four experiments. For each experiment, the sums of the positive and negative increments are given, together with their total. Values are expressed in 10⁻² TgS. A dash line indicates that no observation was assimilated at this time.

surface concentrations of O₃, SO₂, CO, NO, NO₂, PM₁₀, and PM_{2.5}. The metadata associated with the observations describe both the type of station and the environmental conditions under which the measurements are performed. Stations were classified by type as traffic, industrial, or background, and by location as urban, suburban, or rural. Observations are available on the EEA web application (<https://eeadmz1-downloads-webapp.azurewebsites.net/>, last access: 29 January 2026).

Line 90: It is probably beyond the scope of this study, but how is the “case of polluted scenarios” determined from the observations? For each pixels, the total columns are computed using the four profiles. Users have to choose the product they want to use.

Line 178: Why are you not using IFS fields to drive the model, consistent with the chemical boundary conditions in the previous paragraph?

In the operation mode, MOCAGE uses ARPEGE forcings in the global domain and IFS forcing in the regional domain, because MOCAGE is part of the CAMS ensemble. In our study, we want to have similar configuration than the operational one. In the operation configuration, the regional domain of MOCAGE is usually forced by the chemical boundary conditions IFS-COMPO. For the case of the eruption in the Reykjanes Peninsula, the regional domain is forced by the global domain because when MOCAGE is forced by chemical boundary conditions from IFS-COMPO, it leads to an overly extended plume.

We specified that the IFS meteorological forcing is used for the regional domain.

Some meteorological fields, such as wind speed and direction, temperature, humidity, pressure, rain, and clouds, are required because MOCAGE is an offline model. These fields are provided by different numerical weather prediction (NWP) models, Action de Recherche Petite Echelle Grande Echelle (ARPEGE) (Courtier, 1991; Bouyssel et al., 2022) for the global domain and by IFS for the regional domain.

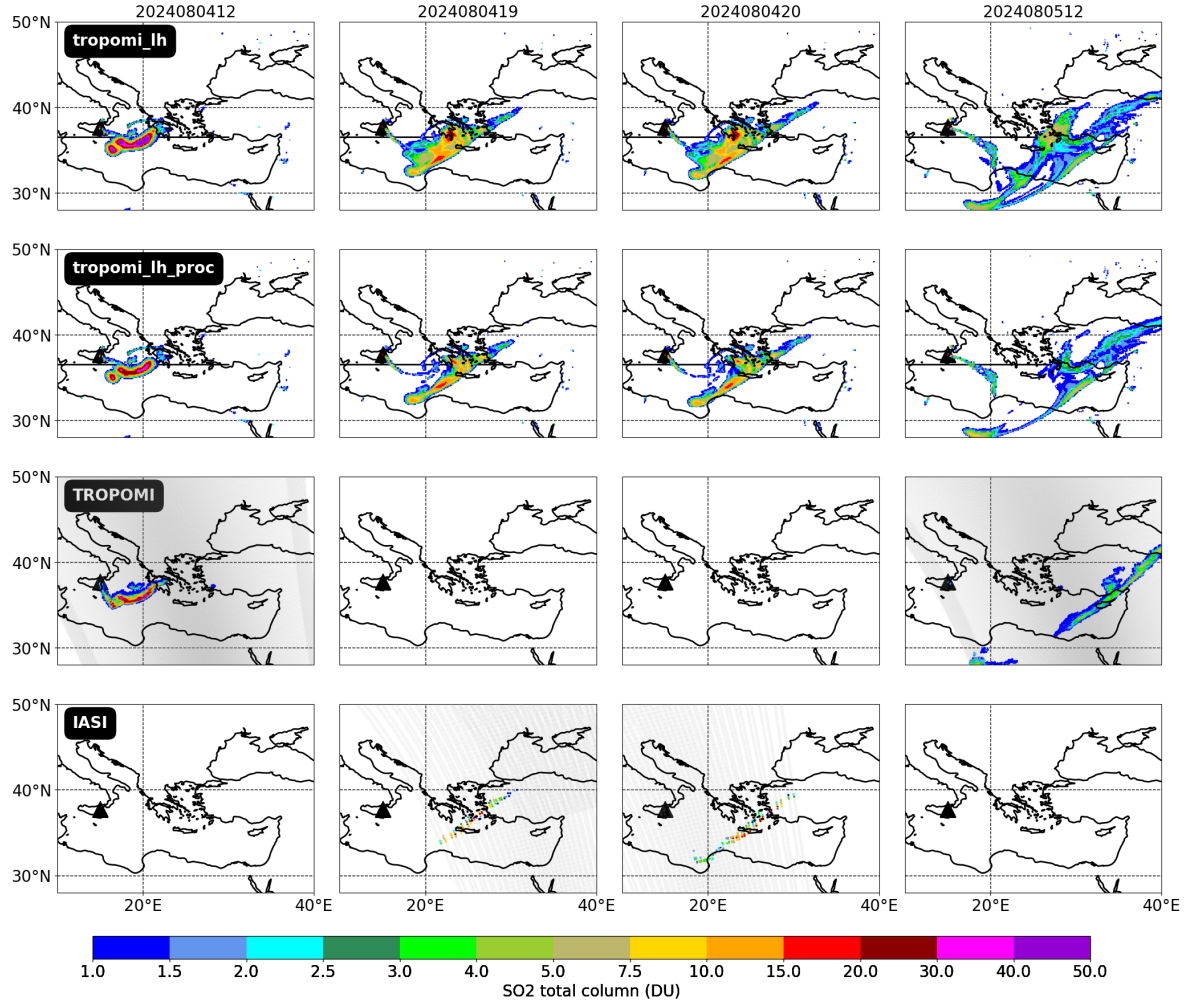
Line 294: What is the rationale for the approach in this algorithm? It seems to aggressively elevate points that are at reasonable altitudes (8-9 km in Figure 3c). Does this make sense?

Assimilating the TROPOMI LH product without observation preprocessing leads to a simulated plume shape that differs from the observed one, particularly on 4 August at 19:00 and 20:00 UTC and on 5 August at 12:00 UTC (FigRev.1). The preprocessed observations allow the model to simulate a plume shape that is more consistent with the observations.

We agree that the altitude increase visible in Figure 3c is locally quite substantial, and that our algorithm has limitations. However, it removes low altitudes at the edge of the plume and also brings the diagnosed altitudes closer to those retrieved by IASI, as shown in FigRev.2.

FigRev.1 has been added to the appendices and the first paragraph of Section 6 has been modified:

For TROPOMI observations, despite the use of quality indicators, the plume altitudes observed at the edge of the plume can differ significantly from those observed at its centre. This discrepancy can be attributed to the partial mixing of clear air and plume, resulting in effective altitudes that are not representative of the main vertical structure of the plume. This behaviour is illustrated in Figure 3 c, where lower plume altitudes, sometimes below 3 km, are observed at the edges of the plume. Assimilating these observations can lead to incorrect plume altitude representation in the model. Due to this incorrect plume altitude representation, the plume can have a different shape than the observed one (FigRev.1).

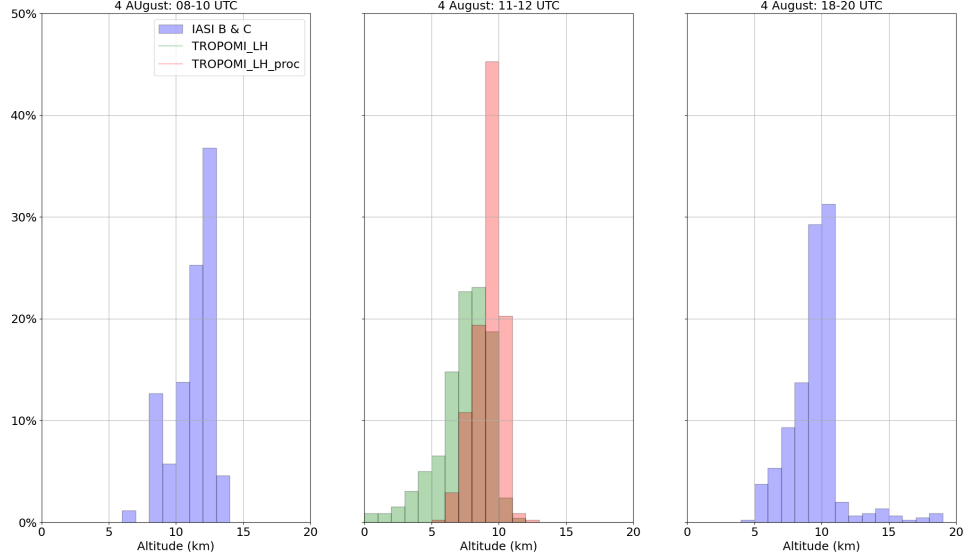


FigRev.1: Analyses of SO₂ total columns on 4 August 2024 at 12:00, 19:00, and 20:00 UTC and on 5 August 2024 at 12:00 UTC. The first two rows correspond respectively to the outputs of the assimilation of raw TROPOMI LH observations and preprocessed TROPOMI LH observations. Observations from TROPOMI and IASI are plotted in the two last row.

FigRev.2 has been added to the paper:

Figure 3 displays the TROPOMI LH observations before (a, c) and after (b, d) the application of the previously described algorithm. The algorithm removes several isolated observations near Greece and increases the plume altitude, particularly around 19°E/36°N and at the plume edges. FigRev.2 shows the distribution of plume altitudes diagnosed by IASI B and C (in blue) for three time windows

on 4 August (08:00–10:00, 11:00–12:00, and 18:00–20:00 UTC), together with the TROPOMI LH altitude distribution before (in green) and after (in red) preprocessing, for the 11:00–12:00 UTC window corresponding to the TROPOMI overpass. Before preprocessing, the TROPOMI LH distribution is broader and centered at a lower altitude than the IASI distributions observed before and after. After preprocessing, it narrows and shifts toward higher altitudes, becoming more consistent with the altitudes diagnosed by IASI. In the following, the observations shown in Figures 3 b and d are assimilated.



FigRev.2: Distribution of plume altitudes (as a percentage of observations within each 1 km bin) for IASI B and C (blue), TROPOMI LH before preprocessing (green) and TROPOMI LH after preprocessing (TROPOMI_LH_proc, red), for three time windows on 4 August 2024: 08:00–10:00 UTC (left), 11:00–12:00 UTC (middle), and 18:00–20:00 UTC (right).

Line 310: You explain why you don't want to use large horizontal correlation lengths, but I wonder if there is an intermediate length that could be tried. The interpolation approach to me seems to invent a lot of data where there is none, which I find problematic. I just can't tell that the high SO₂ going into Turkey is real in Figure 6b. During this study, assimilation was tested using both raw and processed IASI observations. When observations are unprocessed, they may be separated by several grid cells, especially in the regional domain. One way to spread the information derived from the observations is to increase the horizontal correlation length, at the risk of producing an overly spread plume. Another approach is to process the observations to obtain one observation per model grid cell.

FigRev.3 shows model outputs when IASI observations are assimilated. In the first three rows, raw IASI observations are assimilated while varying the horizontal correlation lengths in the background error description: 0.15°, and 0.25°. The fourth row shows the results of assimilating interpolated observations using a fixed correlation length of 0.15°. Finally, the last row shows IASI observations from 4 August at 09:00, 19:00, and 20:00 UTC, and from 5 August at 08:00 UTC.

Using a 0.15° correlation lengths leads to a plume whose the shape is consistent with observations but its intensity remains too low. Using a 0.25° correlation lengths leads to a plume whose the intensity is consistent with observations but it too spread out. Moreover, the correlation length cannot be tuned independently for each instrument: MOCAGE uses a single correlation length per control variable, and SO₂ is the control variable for both IASI and TROPOMI LH. Increasing the length to compensate for the sparse IASI sampling would therefore also spread the TROPOMI LH increments and degrade their assimilation, especially in the joint assimilation experiment. Sensitivity tests for the correlation

lengths for the TROPOMI LH assimilation show that the use of a 0.25° correlation lengths degrade the assimilation (FigRev.4)

FigRev.3 has been added to an appendix and the second paragraph of the Subsection 6.2 has been modified:

The resolution of the IASI pixel is variable. At the nadir, the pixels are a circle of 12 km in diameter, whereas at the edge of the swath, the pixels are an ellipse of around 40x20 km. Nevertheless, MOCAGE has a horizontal resolution of 0.1° (approximately 10 km). Consequently, some IASI pixels cover several MOCAGE meshgrids. This behaviour is shown in Figure 6 representing the IASI pixels on the MOCAGE grid on 4 August 2024 at 20 UTC. This plot also shows some pixels can be separated by several meshgrids, in particular, when the viewing angle increases. The assimilation of these observations without treatment leads to an underestimation of the SO_2 total columns of MOCAGE.

Two approaches can be used to assimilate these observations. The first involves increasing the horizontal correlation length to propagate information from each pixel to neighbouring grid cells. Sensitivity tests (FigRev.3) showed that an intermediate correlation length (0.15°) yields a plume with a shape matching the observations but an underestimated intensity, while a larger length (0.25°) yields a plume with an excessive spatial extent. Moreover, in MOCAGE, a single correlation length is defined for each control variable. Since SO_2 is the control variable for both IASI and TROPOMI LH, a length large enough to spread IASI information would also spread the information from TROPOMI LH, whose pixels are smaller and contiguous, and would degrade its assimilation (FigRev.4). The second approach involves interpolating both the SO_2 total columns and the plume altitudes onto the MOCAGE grid. This approach, which yields the best agreement with IASI observations while keeping a correlation length suited to TROPOMI LH, is the one adopted in this study.

FigRev.4 has been added to an appendix:

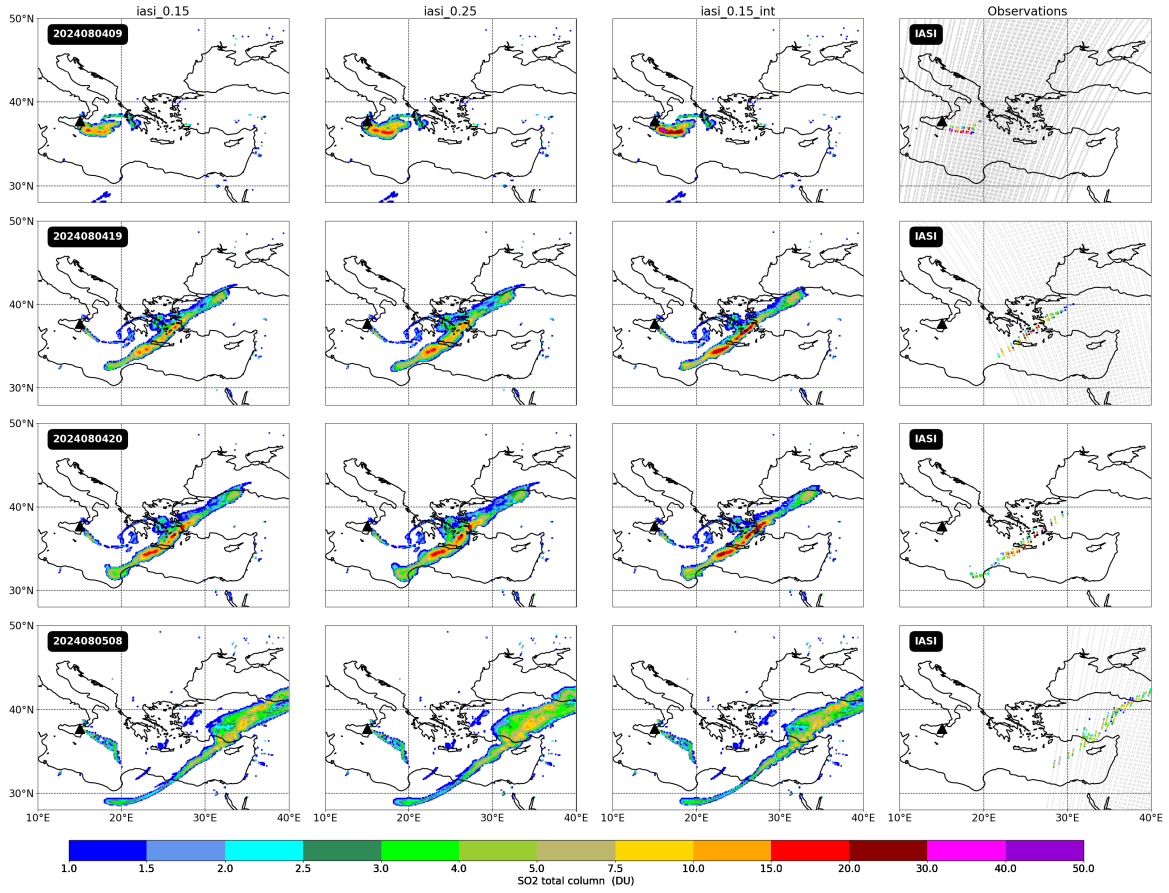
We agree that the interpolation fills gaps between IASI pixels that are not directly observed. This mainly occurs at the edge of the swath, where the scanlines are separated by more than 100 km, as in Figure 6b between 28°E and 30°E . Elsewhere, the interpolated values are consistent with neighbouring pixels: linear interpolation cannot produce values outside the range of the surrounding observations.

However, the presence of SO_2 over Turkey is supported by several observations. Non-zero IASI pixels are present in the raw observations at 20:00 UTC on 4 August (Figure 6a). The next IASI overpasses on 5 August at 08:00 UTC show the plume extending over Turkey (FigRev.3, last row), and TROPOMI observations on 5 August at 12:00 UTC also show the plume extending towards Turkey. In addition, the experiments assimilating raw IASI observations, without interpolation, also produce SO_2 in this region (FigRev.3).

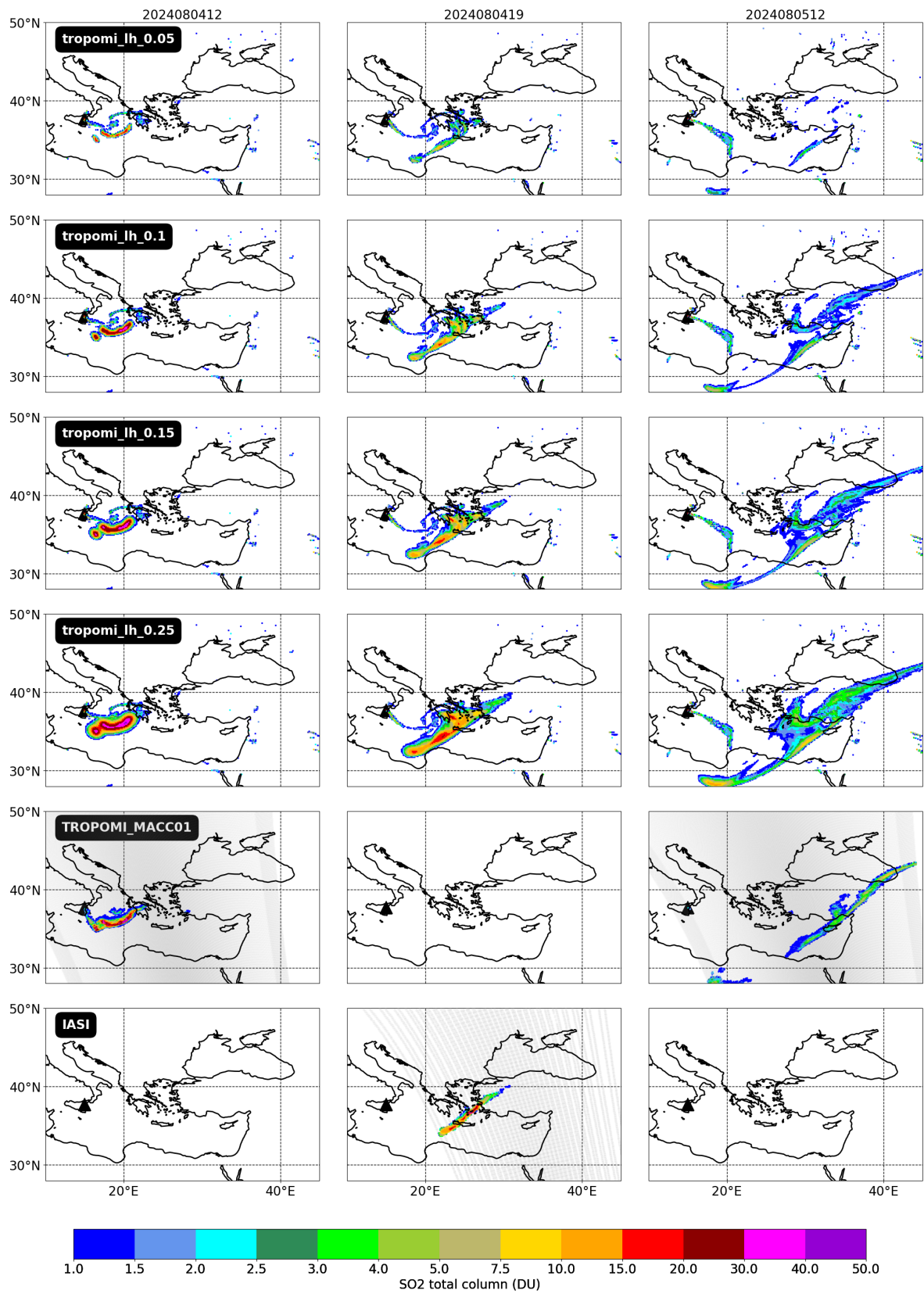
Line 422: There seems to be a real data retention issue in the model. What is happening to this SO_2 ? Is it being scavenged? Rapidly converted to sulfate? How does MOCAGE compare to other aerosol models in this respect? Too aggressive? The sharp decreases shown in Figure 10 do not correspond to a loss of SO_2 in the model. The plume area and SO_2 burden in the figure are computed only over grid cells where SO_2 total columns exceed a threshold (3 or 10 DU).

To clarify this point, we computed the SO_2 and sulfate burden without threshold over the whole regional domain, expressed in TgS (FigRep.1). The volcanic contributions are obtained by subtracting a simulation without assimilation, which includes the same anthropogenic emissions, passive degassing, and meteorological forcing.

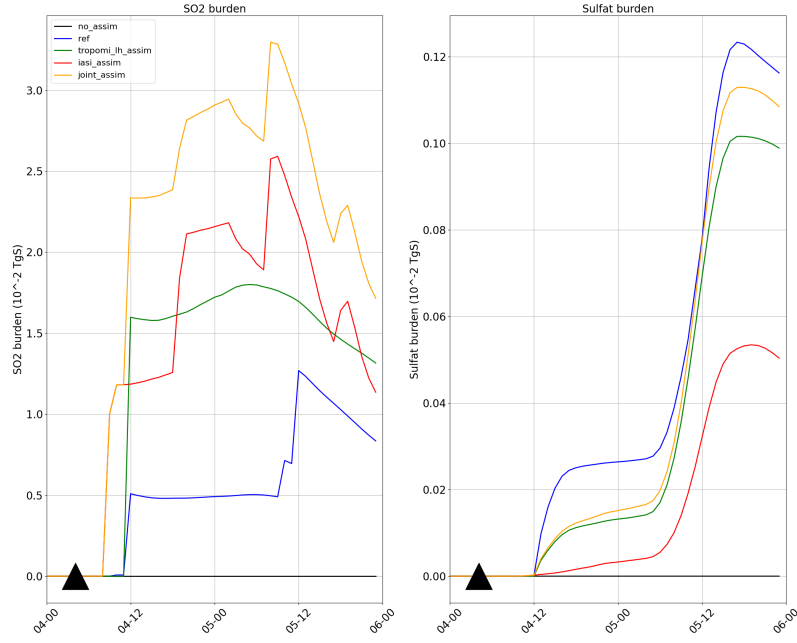
Between two assimilations on 4 August, the volcanic SO_2 remains stable in all experiments. It shows that the sharp decreases in Figure 10 result from the dispersion of the plume below the thresholds rather than from SO_2 removal. The decrease shown in FigRep.1 on 5 August is mainly due to the plume leaving the domain. Sulfate aerosols production remains low.



Figrev.3: Analyses of SO₂ total columns on 4 August 2024 at 09:00, 19:00, and 20:00 UTC and on 5 August 2024 at 08:00 UTC. The first four rows correspond respectively to the outputs of the assimilation of IASI observations by using raw observations and an horizontal correlation of 0.05°, 0.15° and 0.25°, and by assimilating interpolated observations and by using an horizontal correlation value of 0.15°. Observations from IASI are plotted in the last row. Overpasses of satellites are plotted in light grey.



FigRev.4: SO₂ total columns simulating on 4 August at 12:00 and 19:00 UTC and on 5 August at 12:00 UTC by assimilating TROPOMI LH product and by using different values as the horizontal correlation length (0.05°, 0.1°, 0.15°, and 0.25°). The two last rows show the TROPOMI and IASI observations.



FigRep.1: Contributions of the SO₂ data assimilation to the SO₂ and sulfate burden.

We have added some precision to explain the decrease of the plume area and SO₂ burden in the paper:

For the 3 DU threshold, the assimilation of IASI data produces a larger plume area compared to the one simulated in the tropomi_lh_assim simulation, in particular from the midpoint of the studied period. Except for the morning of 4 August, the plume area and the SO₂ burden are significantly larger than the observations. Each IASI assimilation increases both the plume area and the SO₂ burden, although the values of these metrics are already overestimated relative to the observations. For the 10 DU threshold, both the plume area and SO₂ burden are consistent with or slightly higher than the observations. Between each IASI assimilation on 4 August, the plume area and the SO₂ burden decline sharply. This decrease is mainly due to the horizontal and vertical dispersion of the plume, which reduces total columns below the threshold.

Line 430: Maybe, but as noted you seem to also have data retention issues so I'm skeptical. What are your analysis increments? Suggest you add a plot that shows that. As shown in our response to the comment on line 422, the SO₂ added by the assimilation is not rapidly removed in the model: the decreases in Figure 10 are due to the thresholds, while the total volcanic SO₂ burden remains stable between assimilations (FigRep.1). SO₂ added at one analysis can therefore persist until the next one.

FigRev.4 shows the analysed SO₂ concentration together with the corresponding analysis increment (analysis minus background) for the joint_assim experiment during the Mount Etna eruption, at 09:00, 12:00 and 19:00 UTC on 4 August. At 09:00 UTC, the positive increment is co-located with the plume (first IASI overpass), adding SO₂ within the corresponding 2 km-thick layer at the diagnosed altitude. At 12:00 UTC, the increment is located at a lower altitude than the simulated plume, resulting in a thickening of the plume rather than a simple reinforcement of its existing structure. At 19:00 UTC, both positive and negative increments are present, illustrating a horizontal repositioning of the plume: negative increments remove SO₂ while positive increments introduce it. This figure and the corresponding discussion have been added to Section 7.1 of the revised manuscript.

Differences in the shape of the plume between the assimilation of each instrument are mainly due to differences in retrieval altitude. Figure 9 shows vertical cross sections at 36.55°N on 4 August at 09:00, 12:00, 18:00 and 19:00 UTC for the simulation in which TROPOMI, TROPOMI LH, IASI,

and both IASI and TROPOMI LH are assimilated. The last row of the plot displays the plume altitudes between 35.45°N and 35.65°N observed with IASI in blue and TROPOMI LH in red.

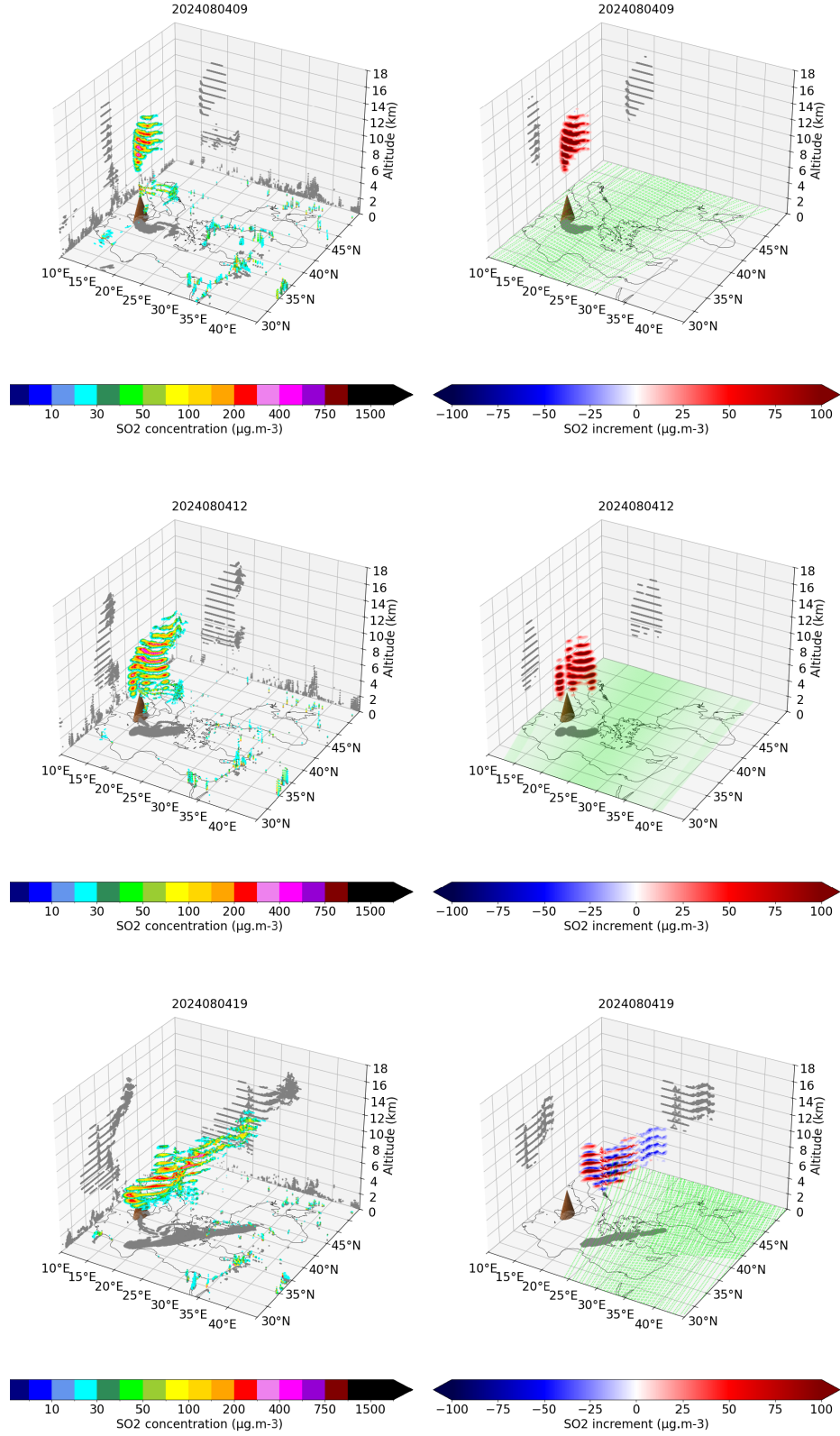
The assimilation of IASI leads to the simulation of a SO₂ plume located between 7 and 15 km of altitude at 9:00 UTC. At the same time, IASI observations detect a plume between 10 and 14 km of altitude. At 12:00 UTC, the simulation that assimilates IASI data shows a plume located between 10 and 14 km of altitude. The modelled plume is higher than that observed by TROPOMI LH, which is located between 7 and 11 km altitude. Due to this altitude discrepancy, the plume in the simulation assimilating TROPOMI LH data is located between 5 and 12 km of altitude. In the joint assimilation experiment, the plume is vertically more extended with a plume located between 5 and 14 km of altitude. In the ref experiment, the assimilation of TROPOMI leads to a simulated plume between 3 and 10 km of altitude around 20°E and 15°E. The simulated concentrations are weaker in this simulation compared to those where altitudes are explicitly considered. The plume simulated around 15°E is not reproduced in the simulation assimilating TROPOMI LH, as the observations do not reach 20 DU. At 18:00 UTC, high concentrations are simulated around 25°E at an altitude of approximately 10 km when IASI data are assimilated. In the other experiments, the plume is more spread out, both vertically and horizontally. The assimilation of TROPOMI LH results in the highest simulated SO₂ concentrations. Simulations assimilating the TROPOMI LH and both IASI and TROPOMI LH instruments yield similar results. The assimilation of TROPOMI produces the lowest concentrations, but the plume structure remains consistent with the experiment assimilating TROPOMI LH.

To better illustrate the assimilation process and its impact on SO₂ concentrations, FigRev.1 shows the analyzed SO₂ concentration and the corresponding analysis increment for the joint_assim experiment at 09:00, 12:00, and 19:00 UTC on 4 August. Before 09:00 UTC, no plume is simulated by MOCAGE. At 09:00 UTC, IASI observations are assimilated, allowing SO₂ to be added to the model between altitudes of 7 and 15 km, as indicated by the positive increments. A plume is thus present within this altitude range. At 12:00 UTC, SO₂ is added at the altitude diagnosed by TROPOMI LH, between 4 and 12 km. However, positive increments occur at a lower altitude than the simulated plume, thanks to the IASI assimilation. Consequently, the thickness of the plume becomes larger. At 19:00 UTC, both positive and negative increments are present, illustrating a horizontal repositioning of the plume: negative increments remove SO₂, while positive increments add it.

Line 455: What dates are being shown in Figure 11? The times do not clearly correspond to, e.g., what is shown in Figure 7. Oh, I see, this is confusing. 04AM means morning on the 4th, etc. To avoid this confusion, we have changed the labels of figures 13 and 14. "04AM" becomes "04-08:AM", "04PM": "04-08:PM"

Line 496: The opening sentence seems not correct. Yes is higher than IASI on 04AM, Tropomi.lh is lest on 05AM. What do you mean to say? We agree that this sentence was not correct. For the 3 DU threshold, the experiments that take the plume altitude into account do not systematically yield higher POD values than the ref experiment. On the morning of 4 August, the POD is lower in the iasi_assim experiment than in the ref experiment, and on the morning of 5 August, the POD is much lower in the tropomi.lh_assim experiment, since no TROPOMI LH observations are available to constrain the model. The paragraph has been modified as follows:

For the 3 DU threshold, the highest POD values are obtained in experiments taking the plume altitude into account, although not systematically. On 4 August, the POD reaches approximately 0.95 in joint_assim experiment, compared to 0.7 in the ref experiment. The joint assimilation of IASI and TROPOMI LH generally produces the highest POD values. On 5 August, the experiment assimilating only TROPOMI LH data shows a lower POD, as no observations were available to constrain the model. For the 10 DU threshold, strong POD values are observed on 4 August in experiments where TROPOMI LH observations are assimilated while low POD are recorded in the other experiments. On 5 August, MOCAGE fails to simulate any SO₂ total columns exceeding 10 DU, whereas observations reach this threshold. For the 30 DU threshold, non-zero POD values are only observed on 4 August in the experiment assimilating TROPOMI LH data. In other experiments, MOCAGE underestimates the total columns, simulating values below 30 DU even when OMPS observes columns of this intensity. On 5 August, no total columns exceeding 30 DU are detected by OMPS. Computed against OMPS



FigRev.4: SO₂ concentration obtained after the assimilation (left column) and analysis increment (right column, analysis minus background) for each model grid point for the joint_assim experiment during the Mount Etna eruption, at 09:00, 12:00, and 19:00 UTC on 4 August 2024. Grey dots show the projection of the plume on the borders of the plot. The green pixels represent satellite overpass.

observations, POD are improved by taking into account the plume altitude in the assimilation. This increase in POD is especially shown for the 10 DU and 30 DU thresholds.

Line 580: Caption to Figure 15 mentions other colored lines which I don't see and don't appear to be described in the text. [We have corrected the caption:](#)

Surface SO₂ concentrations observed (in black) and modelled by assimilating TROPOMI LH (in red). Timeseries are plotted for 24 and 25 August 2024 in Portlaoise (Ireland), in Leeds (United Kingdom), in Calais (France) and in Copenhagen (Denmark).