Dear Editor,

We are submitting a revised version of our manuscript, "Strength of TROPOMI Satellite Observations in Retrieving Hourly Resolved Sources of Volcanic Sulfur Dioxide by Inverse Modeling." We appreciate the two reviewers' constructive comments, thoughtful and insightful critiques that have strengthened our inverse modeling study on the 2018 Ambrym eruption. We thank them for their valuable time invested in correcting our manuscript and streamlining its presentation. We have done our best to address the reviewers' concerns comprehensively in this revision.

We have made the following improvements: the abstract and research method section have been updated; and new analyses have been incorporated. All the figures have been modified for improved accessibility to colorblind readers, and the text has been revised to more effectively prove the strengths of high-spatial-resolution SO2 measurements from TROPOMI in the context of inverse modeling. Although not all comments achieved universal agreement, they proved valuable.

A detailed point-by-point response to the reviewers' comments is provided below, with reviewers' questions and remarks in italics and our corresponding responses in regular font. Line numbers in the draft serve their intended purpose by denoting reviewers' comments from the submitted version. However, in our responses, our references to line numbers refer to the revised version. Recent text additions to the draft are denoted in a distinct color (blue) to differentiate them from the original content (in the first version), which has been redacted using strikethrough formatting.

Reviewer – 1

Major remarks:

1. *However, SO2 mass should not be lost in this smoothing process, so I would expect to see a lower but broader peak in the emissions from OMPS compared to the high but narrow peak from TROPOMI, such as seen when smoothing the TROPOMI time series in figure 7.*

We acknowledge your observation about the impact of spatial smoothing on TROPOMI measurements which should not imply any SO2 mass loss. Supplementary figures (Figs. S26-S31) have been included to illustrate this point. Encountering swath edges with Nan values during spatial smoothing can lead to a minor, second-order loss of SO2 mass. However,

December 17 (Fig. 30) and 18 (Fig. S31) serve as prime examples where spatial smoothing demonstrably does not result in significant SO2 mass loss.

On December 15 (Fig. S28) and 16 (Fig. S29), a distinct phenomenon appears (3D-effect) due to TROPOMI's high spatial resolution measurements (please refer to Wagner et al., (2023) for the details). The SO2 mass within the designated rectangular box shows a clear discrepancy between TROPOMI and OMPS data on these specific days. Signal saturation in OMPS, potentially hindering the detection of dense SO2 plumes (Fig. S12), offers a plausible explanation for this observation. We reference the work of Wagner et al. (2023) to provide further context on the behavior of high spatial resolution SO2 measurements.

Line 399-405: "Moreover, it is imperative to observe that the cumulative mass of SO2 within this defined rectangular area exhibits minimal variation (Fig. 4iii-2, 4iv-2, 4v-2), fulfilling the conservation of SO2 mass expected when performing spatial smoothing (Fig. S26-S31). Nonetheless, it consistently registers approximately 10 kt higher in SO2 mass than OMPS measurements. This intriguing behavior underscores an additional phenomenon atop the high-spatial resolution data provided by TROPOMI. When the concentration of the plume reaches exceedingly elevated levels near its source, it is plausible that OMPS experiences a signal saturation effect (Wagner et al., 2023), consequently leading to an underestimation of the SO2 mass."

Regarding Fig. 7, we have incorporated your valuable suggestion. By assimilating TROPOMI SO2 data smoothed with a disc of radius 75 km, we show that high spatial resolution measurements indeed yield high temporal SO2 flux information via inverse modeling. The substantial absence of very dense of SO2 columns in the OMPS measurements for December 16 manifests as a trough in the SO2 flux time series compared to the results obtained through TROPOMI SO2 data assimilation.

2. *The pre-eruptive SO2 emission is constrained by a few pixels detected in the edge of the swath on both 14th and 15th December. However, what is not clear to me is why the main SO2 plumes seen closer to the volcano on 13th and 14thDecember do not result in any SO2 emission in the reconstructed time series, when these few scattered pixels on the swath edge do? These emissions are also injected at very high altitude in the reconstructed emissions.*

On December 14, our inverse modeling study retrieved SO2 injection heights (#T1 emission at ~11 km; Fig. 3) reveals an eastward-migrating plume detected by TROPOMI at the swath edge (Fig. S14). This eastward movement is primarily attributed to the SO2 injection at remarkably high altitude (Fig. S32). Based on external calculations, HYSPLIT trajectory model using the 0.5˚ GDAS reanalysis are provided (supplementary material) for complementary validation of our inverse modeling results. In executing the HYSPLIT trajectory model, we

engage the web interface furnished by NOAA (https://www.ready.noaa.gov/hypub-bin/trajtype.pl), whence we retrieve the computations and graphical outputs. These computations are conducted on their server.

Westward movement seen on December 13 and 14 (Fig. S32-ii) originates from SO2 injection at a lower altitude (1.3 km ASL). Sensitivity of polar-orbiting sensors is generally limited towards such low-level plumes, as evidenced by the averaging kernel vertical profiles (Fig. S18). Hence, the column values of SO2 would inherently retain significant uncertainty due to the use of a particular SO2 product variant (in this case, the 7 km SO2 product). The western plume in question is still unaccounted for in our inverse modeling procedure, a consequence of its nonappearance in modeled parameters due to the constraints of the averaging kernels (Eskes et al., 2003) in detecting these low-altitude emissions via polar-orbiting satellites.

3. *In figure 6, the authors compare the observed and modeled plumes for each sensor on three days, marking areas of cloud cover. However, it is not clear how these areas of cloud are defined.*

For the sake of presentational clarity, the initial figure was constructed visually, omitting contour lines. As you suggested, we have now generated a revised version incorporating these features to depict 0.3 cloud fraction values. The high spatial resolution of TROPOMI data, however, results in a comparatively intricate network of contour lines. Conversely, the OMPS measurements yield a smoother presentation due to their coarse spatial resolution.

4. *What is not clear to me is physically how this SO2 is transported from the dyke to the surface, and where it is emitted? This section requires further explanation on the mechanisms of this release.*

To ensure clarity and address your inquiries, we have undertaken a revision of sect. 4.3 to explain the mechanism of gas/magma separation at the dike tip that is envisaged for the Ambrym case-study (see details in the text below). Additionally, a new figure has been incorporated to illustrate the temporal evolution of degassing in comparison with other geophysical observations (ground deformation, seismicity, and thermal activity). Furthermore, citations to other pertinent studies have been included to support the robustness of our explanations. We have included the following text to address your specific query.

Section 4.3: "Following an initial moderately-sized intrusion of magma (34×10^6 m3, Shreve et al. (2019)), which instigated the eruption late on December 14, draining active lava lakes and producing in an intra-caldera lava flow reaching its maximum extent by late the following day, a significantly larger dike (419×10^6 m3 to 532×10^6 m3, Shreve et al. (2019)) was later intruded on December 15 at a shallow depth. This second dike then propagated laterally into

the rift zone outside the caldera (Shreve et al., 2019) and caused a substantial ground deformation of the island. The ensuing ground deformation of the island was with coastal uplift exceeding 2 m, as thoroughly documented by Shreve et al. (2019, 2022). The dike's extension towards the eastern region of the island caused surface ruptures, culminating in an offshore submarine eruption.

In the initial stage of the eruption, a synchronous progression emerges between the temporal dynamics of thermal activity of the lava flow, as illustrated by the thermal index of lava flow pixels obtained from Himawari-8/AHI observations (green profile in Fig. 8c), and the temporal evolution of SO2 flux derived from TROPOMI data (red profile in Fig. 8c). Notably, SO2 emissions labeled #T2 and #T3 align with a surge in lava flow activity on December 15, spanning from 00:30 to 04:30 UT, as documented by Shreve et al. (2019). Emission #T2 ascends to altitudes ranging between 9 km to 10 km ASL, expelling approximately 3.8 kt of SO2. Simultaneously, emission #T3 expels about 2.75 kt of SO2 at an altitude of 5 km, accompanied by an additional 0.5 kt at 2 km ASL. Antecedent to a notable seismic volcano-tectonic occurrence around 20:00 UT on December 15 (Shreve et al.,2019), emissions labeled #T4 and #T5 likewise coincide with the emplacement of the lava flow, suggesting that the primary source of SO2 degassing in this phase is the magma feeding the expanding intra-caldera lava flow (Fig. 8c).

Upon the advent of the second stage of the eruption on December 16, a discernible shift in eruption dynamics becomes apparent. During this interval, the thermal activity within the intra-caldera lava flow regresses to a state akin to the pre-eruptive baseline, denoting the cessation of lava emission. This deduction, corroborated by the analysis of Sentinel-2 imagery as documented in Shreve et al. (2019), suggests that the lava flow has reached its maximum extent. Yet, a momentous surge in SO2 emissions is unveiled by the TROPOMI-derived SO2 flux data, with a sudden escalation noted at emissions #T7 and #T8. Over 10 kt of SO2 is discharged, ascending to altitudes of approximately 11 km ASL (Fig. 3a, S34). This augmentation corresponds to the most notable paroxysmal degassing event since the onset of the eruption. It occurs in the wake of renewed seismic activity, as documented by Shreve et al. (2022), and coincides with the initiation of seismicity migration from the caldera towards the eastern periphery of the island (Fig. 8b). This seismic migration offers direct evidence of the lateral propagation of a voluminous dike traversing the upper crust from the island's central region into the eastern rift zone. On the other hand, the subsequent lateral progression of the magma does not seem to align with a substantial gas release at the surface.

Theoretical investigations (Lister, 1990; Rubin, 1993), later substantiated through experimental studies (Menand and Tait, 2001; Maimon et al., 2012), have illuminated the phenomenon wherein volatiles exsolve from magma, birthing a gas pocket at the dike tip. Such gas pockets may detach from the magma-laden fissure and ascend towards the surface, manifesting as a disconnected crack brimming with gases. The marked surge in gas emissions during the early hours of December 16 coincides precisely with the initiation of seismic migration (Figs. 8a, 8b) when the dike tip has recently vacated the caldera but is yet to reach its full lateral extent, a nuance perceptible through sub-pixel correlations of Sentinel-2 optical observations (Shreve et al., 2019).

Our analysis suggests that the heightened degassing observed on the early hours of December 16 likely stems from gases originating from the substantial dike intrusion. Gas pockets detached from the magma while the dike rose from the central reservoir situated at depths exceeding 4 km (Shreve et al., 2022; Moussallam et al., 2021) to subsurface levels a few kilometers in depth (Shreve et al., 2019). These gas parcels, after their detachment from the melt rock, presumably followed the most direct route to the surface, likely leading to pre-existing intra-caldera vents opened by the preceding dike, such as Lewolembwi and other vents on the southeastern flank of Marum (Fig. 8). The precise thickness of the magma intrusions from December 14 to 15 is unknown, raising the possibility that feeder fractures had not yet solidified by December 16. Moreover, while the exact point of gas emission stays elusive, we have not detected any substantial shift in the location of SO2 emissions from the commencement of the eruption through early December 16. This observation is based on high-resolution Himawari-8/AHI SO2 RGB images, boasting a native resolution of 4 km at the sub-satellite point, marginally oversampled to 3.5 km (see animation M1, Figs. 5, S6). Such findings support the occurrence of a substantial intra-caldera SO2 gas release linked with the initiation of dike propagation, a phenomenon that might have otherwise gone unnoticed in the absence of a relatively open system.

This conclusion seems to contradict the findings of Shreve et al. (2019), who posited, through a rough comparison of SO2 mass and lava flow volume, that the majority of the SO2 budget should be ascribed to the lava flow, with the lateral dike's contribution to degassing being minimal. Our investigation offers a more nuanced perspective, suggesting that a part of the magma injected into the lateral dike could have contributed to the pronounced degassing peak observed at the onset of its lateral migration, potentially as the magma transitions from an initially vertical to subsequently lateral migration. However, this notable peak in SO2 degassing, occurring in early December 16, accounts for less than 20% of the total SO2 release, reaffirming that the lava flow on December 14–15 is still the principal source of degassing. Thus, the conclusion drawn by Shreve et al. (2019) holds valid at the first order.

Our interpretations suggest that the lateral migration of dikes along the rift does not result in discernible SO2 emissions above the body of the dike in lateral motion; in other words, no SO2 release is observed from the fractures situated above the laterally migrating dike. Likewise, for instance, although weak satellite detection of SO2 is associated, degassing from the volcanism of the Ethiopian rift has primarily been linked to the erupted lava rather than the loss of volatiles from unerupted magma dikes (Ferguson et al., 2010; Barnie et al., 2016).

Thus, the assimilation of TROPOMI SO2 column observations facilitates the extraction of altitude-resolved SO2 emissions with hourly resolution. This procedure not only supports the tracking of rapid changes in eruptive dynamics but also helps in discerning diverse magma sources of SO2 discharge. Furthermore, it presents an advantage over Himawari-8/AHI observations due to its limited susceptibility to ash co-emission interference. The heightened temporal resolution on SO2 emissions provided by the assimilation of LEO satellite observations into inverse modeling could represent a supplementary asset for strengthening the endeavors of volcanological observatories in hazard evaluation."

5. *My final major comment is on a lack of discussion of uncertainty. There are uncertainty estimates for the total masses given (L265 and L506) but there is no discussion on how these errors are derived and no errors given in any other values. I feel the manuscript requires these to be added, both to values in the text and to the figures of the emission time series.*

Firstly, the uncertainty associated with SO2 column retrievals depends on the chosen satellite product. As illustrated in Figs. S20-S25, varying assumptions about the center of mass for SO2 mass concentration led to discrepancies in both total SO2 column and mass across 1 km, 7 km, and 15 km level-2 SO2 products. Given the intermediate nature of the 2018 Ambrym eruption – exceeding neither minor nor stratospheric-reaching major events – the 7 km SO2 product is a prudent selection, aligning with the approach of several published works on this eruption. In the interest of our understanding, we present the results of assimilating SO2 column data from three TROPOMI products (Fig. S33). Notably, the assimilation of the 1 km SO2 product reveals significantly larger mass flux values compared to the other level-2 products. This observation can be attributed to the substantially higher column abundances of SO2 within the 1 km product. It should be noted that the temporal evolution of the SO2 mass flux is the same, whichever the TROPOMI product that is used. There is however a scaling factor approximately 3 (Fig. S33).

Secondly, the vertical sensitivity of the retrieval method diminishes near the surface, showed by a sharp decline in averaging kernel values below 2-3 km ASL. Consequently, the uncertainty associated with weaker, lower-altitude emissions is amplified. To mitigate these uncertainties and potential biases in retrieved SO2 fluxes, we employ a ($\mu+\sigma$) threshold on SO2 source amplitude strength. Sources falling below this threshold, showing the paradoxical combination of weak emissions and unrealistically high injection heights, are excluded from further analysis (detailed in Fig. S5). We have resolved the uncertainty in the total mass of SO2 in TROPOMI and OMPS by cross-referencing Fig. S5, and revisiting section 2.4.2.

Finally, uncertainties inherent to transport model simulations, which incorporate numerous external meteorological variables from archived meteorological (reanalysis) data and WRF simulations, are challenging to relate to retrieved flux values. Therefore, for the sake of clarity, we assume these simulations to be correct and refrain from propagating any associated uncertainties into our results.

Line 353-354: “The determination of the uncertainty in the aggregate mass of SO2 relies on the post-processing phase described in section 2.4.2 (Fig. S5), with additional elaboration provided in sect. 4.2.2.”

Minor/Specific remarks:

1. *L31: "SO_2 is an unambiguous indicator of volcanic plume" I wouldn't necessarily agree with this, as SO_2 plumes from anthropogenic sources are often visible in satellite imagery.*

   Thanks for your remark; we have removed this ambiguity.

2. *L49-50: Assuming the inputs are daily data from LEO sensors, the delta-M method does not provide sub-daily emission rates, I would suggest rewording this.*

   Thanks for your remark; we have rephrased the sentence.

3. *L54: The stacking method does not have to be monthly; it can be whatever time resolution desired.*

Indeed, your observation is correct. However, when discussing the method about the estimation of SO2 mass, emission rates, and the accompanying references, the estimations are conducted at a monthly time scale. Our first assertion stays unchanged.

4. *L56: The delta-M method does not require wind field data to my knowledge.*

We appreciate your feedback, though it is important to note that our assertions are not entirely inaccurate. Referencing Eq. 5 in Theys et al.'s work (ACP, 2013), "k*M(t)" denotes the term for SO2 mass loss within the Delta-M method. While wind fields are not explicitly mentioned in this equation, the efficacy of this mass loss term depends upon factors such as dry and wet deposition, losses during transport, and dilution/diffusion within the plume's periphery. Consequently, all these mechanisms are inherently tied to the influence of wind fields. Thus, we support our original statement with minor modifications.

Line 82-87: "It is essential to recognize that the effectiveness of these methodologies rests upon the assumptions about plume dispersion and wind fields, whether explicitly articulated or implicitly deduced. As such, they might fall short in scenarios characterized by intricate wind patterns. However, of late, there have been endeavors to estimate daily SO2 mass flux from space-borne hyperspectral SO2 column images, aiming to do so without prior knowledge of plume direction or speed. The algorithm devised in this pursuit enables the automatic estimation of SO2 flux alongside its corresponding uncertainty (Grandin et al., 2023)."

5. *L62: Back-trajectory analysis only has issues with re-circulating plumes in some situations with large eruption plumes, and this issue is not unique to this method. In particular, in the paper by Queißer et al. (2019) recirculation is not highlighted as a drawback with this approach, so I would suggest rewording this.*

Thank you for your suggestion, with which we largely concur at this point. The challenges inherent in back-trajectory analysis and the dynamics of re-circulating plumes are indeed amplified with larger plumes. However, in Queißer et al. (2019), the plume length extends to over 400 km, a scale ample to potentially undergo such re-circulatory effects (please refer to Fig. 2b in their publication). This finding appears to present a counterpoint to the assertion made here. As such, we support our original phrasing in this context.

6. *L66: Plume injection altitude can also be used as an a priori.*

   We have revised the statement to include the injection altitude.

7. *L99-103: The discussion of conversion of slant columns to vertical columns needs more information. Specifically, the TM5 profile is not usually used in volcanic applications, this is typically the 1, 7, and 15 km box profiles, which are not mentioned here. Also, the air mass factors computed and contained within the TROPOMI product files include information on the scattering weighting function and reflecting surface, these are not separate parameters. If discussing them separately, I would highlight that it is the geometrical air mass factor that is combined with these other factors.*

Thank you for your valuable insights. We have revised our text concerning the transformation from the slant column to the vertical column density, which is referred as total column in this study.

Line 134-145: "To effectively calculate the comprehensive SO2 vertical column from SCD at each footprint, the algorithm necessitates precise air mass factor data, intrinsically dependent on the wavelength under consideration. This calculation of air mass factors demands a host of ancillary inputs, including but not limited to the computation of scattering weight functions by the radiative transfer model, which operates under the assumption of a U.S. standard atmosphere; the incorporation of a Lambertian reflecting surface to account for meteorological cloud conditions (Eskes and Boersma, 2003); factors such as solar zenith angle and viewing azimuth angle; as well as the information on topographic data for surface pressure, total ozone column, and the structure of the vertical SO2 profile. These a priori shapes of the SO2 vertical profiles are derived from the computations of an external, offline CTM, assuming three distinct scenarios wherein the center of mass of SO2 profile resides at altitudes of 1 km, 7 km, and 15 km above sea level (ASL). Notably, the CTM employed herein is TM5–a tracer model characterized by a spatial resolution of $1^\circ \times 1^\circ$ and encompassing 34 sigma pressure levels, spanning vertically up to 0.1 hPa Huijnen et al. (2010). The TM5 model derives its meteorological input from the operational data of the European Centre for Medium-Range Weather Forecasts (ECMWF)."

Line 160-163: "Much akin to the DOAS algorithm, the PCA algorithm likewise retrieves SO2 column values by assuming the center of mass within the vertical profile of SO2 at various

altitudes—namely, the PBL (1-2 km), TRL (3 km), TRM (7 km), TRU (13 km), and STL (18 km)."

8. *L120: Here you say that you use the 7 km VCD product, however, the plumes you measure are injected at a wide range of altitudes. How do you account for the changing sensitivity to SO2 with altitude? This could dramatically impact the reconstruction results.*

You are correct, and we have indeed included a figure while addressing your fifth major remark (see Fig. S33). The injection of SO2 manifests across a broad spectrum of altitudes, depending upon the scale of the eruption, as exemplified in the 2018 Ambrym event. However, for the satellite level-2 (DOAS) product, we have opted for the 7 km SO2 column product. This choice, maybe, stands as a limitation of our study. Moreover, your inquiry may suggest consideration of assimilating the COBRA SO2 product (Theys et al., ACP, 2021), acknowledged for its heightened accuracy compared to DOAS products. Nonetheless, we have demonstrated that altering the satellite SO2 column products sourced from TROPOMI itself does not impact the timing and injection heights of SO2 emissions. The primary aim of our investigation is to underscore the strength of TROPOMI's high spatial resolution SO2 measurements in inverse modeling, a purpose realized through the utilization of the 7km SO2 product from TROPOMI and the TRM SO2 product from OMPS, both deemed equivalent. The exploration of uncertainties associated with different TROPOMI SO2 products and their outcomes on inverse modeling stands as a prospective avenue for further research.

9. *L122-125: The discussion of the thresholding at the swath edges is not fully clear. You remove any VCD > 1 DU, but then "set a specific threshold for the SO_2 column values of pixels at the swath edge" and manually tune this threshold to 1.1 – 1.4 DU. Does this mean that you discard any pixels that are 1 < VCD < 1.1 (if using a 1.1 DU threshold)? Also, how did tuning this threshold impact the detection of plume in the swath edge attributed to pre-eruptive degassing?*

We appreciate your comment, which prompted us to revise the statement as follows. This adjustment also allows us to reference Figure 2 from the work of Fioletov et al. (ACP, 2020).

Line 170-175: "Before assimilation, we post-process the TROPOMI data to eliminate anomalously high SO2 vertical column values present at the swath edges (see Fig. S1). We define the swath edge as encompassing 25 ground pixels on both sides of the swath (Fioletov et al., 2020). To safeguard against inaccurate retrievals disturbing the inverse modeling procedure, we establish a specific threshold for SO2 vertical column values of pixels at the swath edge, distinguishing the volcanic SO2 plume and discerning noise through visual inspection"

10. *L153-154: Why limit the analysis to above 2 km? There appear to be emissions lower than this, so would it not be better to include these instead of disregarding them?*

Your question regarding this matter has been previously addressed in response to your second major remark, substantiated through the usage of averaging kernel vertical profiles (see Fig. S18). However, the enhanced accuracy and sensitivity of the COBRA SO2 product (Theys et al. 2021) in detecting faint, frequently encountered SO2 plumes at lower altitudes heralds forthcoming avenues of inquiry, wherein the COBRA SO2 product is poised to play a pivotal role in inverse modeling procedures. Supplementary support regarding the use of emission profiles exceeding 2 km ASL has been inserted into the main body of the text.

Line 206-211: "The rationale behind restricting SO2 emission regulations to altitudes spanning from 2 km to 11 km ASL stems from the constrained sensitivity of LEO UV sensors at low altitudes (<2 km ASL), attributable to diminished sensitivity as evidenced by previous studies (e.g., Yang et al., 2010; Theys et al., 2017) (refer to Fig. S18, Eskes and Boersma (2003)). At an elevation of 1.3 km ASL, Ambrym's vent stands, prompting a mindful consideration of the constraints inherent in satellite measurements. Consequently, our scope of investigation narrows to emissions surpassing the 2 km ASL threshold, thus characterizing a more precise focus on the eruptive phase of SO2 degassing. Therefore, we do not expect the CTM simulations to generate an SO2 plume to the northwest of the source, as driven by the low-level southeasterly trade winds. Furthermore, to substantiate our selection of the highest SO2 injection altitude in the CTM simulation, we draw upon insights obtained from IASI SO2 height products (Clarisse et al., 2014), revealing that the primary altitude of the highest SO2 layer predominantly resides within the 10 km to 11 km range ASL (see Fig. S19)."

11. *L236-237: You state "a clear correlation emerges with high values of the lava flow indices". Although there is some similarity (peak at 00:00 on 15th December, both decay from 12:00), the peak in the lava flow proxy coincides with a trough in emissions, so I do not think this sentence is necessarily valid.*

Thank you for your observation, although it is not quite clear. At approximately 00:00 UT, there is concurrence in the peaks; from 04:00 to 06:00 UT, both peaks and troughs align; spanning 06:00 to 12:00 UT, thermal anomalies indicative of lava flow present three peaks, yet inverted SO2 emissions reveal two peaks. Is this the distinction to which you refer? If so, the term "clear" has been omitted from your statement.

12. *L297: "TROPOMI's hyperspectral…" – do you mean here that TROPOMI has a higher spectral resolution (~0.5 vs ~ 1 nm)? I am not sure how much difference the spectral resolution makes, for example TROPOMI has a similar spectral resolution to OMI. The higher sensitivity is driven primarily by the spatial resolution.*

Enhanced by hyperspectral features, which depicts heightened spectral resolution, the discernment of trace gases such as SO2 reaches remarkable sensitivity. This enhancement in

spectral resolution increases the signal content per unit pixel, culminating in an elevated signal-to-noise ratio. Consequently, this advancement facilitates the identification of minute concentrations of SO2 plumes (Theys et al., 2017; Wagner et al., 2023). While a 1 nm spectral resolution suffices for SO2 retrieval, a finer 0.5 nm threshold heightens the detection ability for SO2 (see, for instance, the comparison of OMI and OMPS in Zhang et al., 2017).

13. *L299-309: You show here that by spatially smoothing the TROPOMI data to the spatial resolution of OMPs you can recreate similar VCDs, but is it not the total mass in view that is more important? Smoothing the data out should conserve the total mass in the view, and so the emission rate reconstructed should be the same when integrated with time. Have you tried reconstructing the emission rates with the smoothed TROPOMI data to see if it matches the OMPS time series?*

Thank you for this observation. Although we recorded the total mass of SO2 within the rectangular box (see Fig. 4.2), we inadvertently omitted reference to it in the main text. To rectify this, we have introduced additional sentences emphasizing the significance of the total SO2 mass detected by both sensors and highlighting that spatial smoothing does not influence the total SO2 mass within the specified region (Figs. S30 and S31). TROPOMI's high-spatial resolution SO2 observations reveal another phenomenon. Specifically, when the SO2 concentration is exceptionally high near the source (Figs. S28 and S29), we also acknowledge the findings of Wagner et al. (2023), who reports signal saturation and 3D effects, leading to an increased SO2 mass detected by TROPOMI compared to OMPS due to pixels exhibiting notably high SO2 column values (e.g., Figs. S28 and S29).

Lines 386-389: "Consequently, the cumulative mass of SO2 within the northeasterly inclined rectangular area measures approximately 53.3 kt as ascertained by TROPOMI (Fig. 4ii-2), contrasting with a SO2 mass of merely 43.1 kt captured within the identical region by OMPS observations (Fig. 4i-2)"

Line 399-405: "Moreover, it is imperative to observe that the cumulative mass of SO2 within this defined rectangular area exhibits minimal variation (Fig. 4iii-2, 4iv-2, 4v-2), fulfilling the conservation of SO2 mass expected when performing spatial smoothing (Fig. S26-S31). Nonetheless, it consistently registers approximately 10 kt higher in SO2 mass than OMPS measurements. This intriguing behavior underscores an additional phenomenon atop the high-spatial resolution data provided by TROPOMI. When the concentration of the plume reaches exceedingly elevated levels near its source, it is plausible that OMPS experiences a signal saturation effect (Wagner et al., 2023), consequently leading to an underestimation of the SO2 mass."

14. *L332: Can you expand on the phrase "In contrast, ash remained at lower altitudes due to wind shear"? Wind shear would explain the separation of ash and SO2 if injected at different altitudes, but it would not cause the ash to be at lower altitudes.*

We have revised the sentences for clarity and precision. We wish to explain that, given the persistent vertical wind field, ash, being denser than SO2, is inclined to reside at lower altitudes when co-emitted. This phenomenon is expounded upon in Fig. 5, wherein we illustrate the bifurcation of SO2 injection at distinct altitudes: one at approximately 5 km and another at higher altitudes of around 10-11 km. The presence of co-emitted ash at these lower altitudes is substantiated and corroborated through external computations employing the HYSPLIT trajectory model, alongside analyses of SO2 RGB images sourced from the geostationary Himawari-8.

Line 430-443: "Furthermore, through external Lagrangian model calculations (HYSPLIT trajectory model (Stein et al., 2015)), driven by 0.5-degree GDAS reanalysis, it is demonstrated that the release of SO2 at 00:00 UT on December 15 at an altitude of 5 km ASL (Fig. 5-vii) effectively replicates a specific branch of the plume, characterized by its north-northeast movement. This trajectory aligns closely with observations made by the geostationary Himawari-8/AHI (Fig. 5iv–5vi). The HYSPLIT Lagrangian model (Stein et al., 2015), forced by 0.5-degree GDAS reanalysis, shows that releasing ash particles at 00:00 UT on December 15 at an altitude of 5 km ASL (Fig. 5-vii) reproduces one branch of the plume moving north-northeast, which is likely the ash branch observed by Himawari-8/AHI (Fig.5iv–5vi). Releasing SO2 at higher altitudes (Fig. 5-viii, 5-ix) fits well with the spatial extent and direction of the SO2 plume indicated by the light greenish plume in the SO2 RGB composites from Himawari-8/AHI observations. HYSPLIT simulations hence validate the bimodal SO2 injection during the eruption's commencement, aligning with the observed bifurcation of the plume into distinct branches at varying altitudes. The bimodal SO2 injection at the eruption onset is a crucial finding that underscores the importance of high spatial resolution satellite observations in capturing the initial phase of volcanic SO2 degassing by inverse modeling."

15. *L361: It is not clear here how the high spatial resolution of TROPOMI helps to detect gas below cloud. If I understand correctly, this is achieved by using multiple images over several days such that any blocked plume is visible in other scenes. So OMPS should also show this behavior (and indeed the model results look broadly similar for TROPOMI and OMPS in terms of spatial extent).*

Thank you for this observation. Indeed, through the assimilation of successive daily SO2 maps from both LEO sensors, we are able to discern SO2 plumes beneath cloud cover using inverse modeling procedure. Fortunately, on selected days, the complete SO2 plume is visible, unaffected by cloud cover. This visibility helps the reconstruction of SO2 sources through inverse modeling, enabling the prediction of SO2 dispersion even on overcast days. To avoid potential confusion, we have rephrased the sentences accordingly in sect.3.2.

16. *L366: "yet faint SO2 signals are visible in TROPOMI observations". I am not sure I agree here from the figures shown. By eye, the SO2 in the red regions looks of a similar level to elsewhere in the image outside the plume. Can you show that the level of the*

SO2 VCDs in this region are above the background noise? L366: How are the red contoured areas defined and what is the source of the cloud data? As shown above, the cloud product within the TROPOMI data, which appears to map well with seen gaps in the plume, do not show significant cloud cover for all regions marked.

Fig. 6 has been revised following your major third comment. By using contour lines depicting CCF values of 0.3, it is discerned that OMPS primarily finds SO2 column values mostly below 0.3 DU (see Fig. 6c, 6d), akin to the background noise level (Li et al., 2020). Conversely, in the context of TROPOMI observations, it is noted that these clustered pixels, impeded by clouds, exhibit SO2 column values surpassing 0.3 DU (see Fig. 6ii). To mitigate confusion, we have also revised the sentence accordingly.

Line 476-485: "Figs. 1 and 2 show that the initialization of the CHIMERE CTM simulation with inverted SO2 emissions yields an elongated SO2 plume that extends towards the north-northeast of Ambrym. This phenomenon is primarily seen on December 17 and 18, exhibiting SO2 column values surpassing the background noise levels in both the assimilations. The crux of the matter now pivots to the question: "Does the extended plume depicted in the CTM simulation, observed within the northeastern quadrant of the domain on December 17–18, possess veracity? If so, by what means?" The challenge lies in the detection of this plume by both sensors; nonetheless, subtle SO2 signals—exceeding the background noise levels—are discernible amidst the cloudy expanse, forming clusters distinctly clear in TROPOMI observations (Fig. 6b). Observations of this nature favor the certitude that these signals do not merely constitute extraneous noise artifacts; rather, they signify the presence of volcanic SO2, affected by the cloud cover."

Line 492-496: "The enhanced spatial resolution of TROPOMI substantiates the plume's existence within cloudy regions and facilitates the validation of both our CTM simulations and inverted SO2 sources. This underscores the capability of our inverse modeling procedure to assimilate successive LEO satellite data on a daily cadence."

17. *L371: If OMPS is not able to detect the SO2 below the cloud, then why does the model create emissions in this region? What information is it using to place the plume here that the model with TROPOMI is not?*

We regret any confusion caused. The crux of the matter we aim to highlight here is the assimilation of successive SO2 column maps from both LEO sensors, resulting in an SO2 plume within the model prediction upon initializing the CTM with inverted SO2 sources. The pivotal inquiry becomes: "Does the expanded plume in the model prediction, observed in the northeastern sector of the domain on December 17-18, hold validity? If so, by what means?" Thus, we delve into the values of cloud fraction, revealing that in regions affected by clouds, TROPOMI SO2 column values surpass the background noise level, instilling confidence in our retrieval process. Conversely, this confidence is lacking in the case of OMPS SO2 columns in these areas, often showing values below the established confidence threshold. Subsequently,

we can confirm our retrieved SO2 sources through inverse modeling. We have tried to rephrase these statements (lines 476-485) to enhance clarity.

18. *L392: Why is the 7–8-hour solution picked? By eye, I would argue that the 4-hour solution is a better fit,, I suspect that the better correlation in the 8-hour smoothing is driven primarily by the paroxysmal peak missing in the OMPS time series. Also, the smoothed results appear to be shifted in time (the peak in the green data shifts later the longer the smoothing window applied). Is this an artefact of the smoothing?*

This task has been completed as advised. Following your recommendation, we considered it imperative to assimilate the smoothed TROPOMI data within a disc of 75-km radius, showing a resemblance to the 4-hour simple moving averaged TROPOMI inverted SO2 flux rates, closely aligning with the SO2 flux values derived from assimilating OMPS measurements, as showed by the Pearson correlation coefficients. Additionally, Fig. 7 has been revised for enhanced clarity. Furthermore, this section has been restructured within the discussion.

The anticipated effect of shifting the smoothed time series towards the righthand side is inherent in the application of a larger window size. This phenomenon arises from the principle of Simple Moving Average (SMA), wherein an increase in window size renders the SMA more attuned to longer-term trends. Should the latter part of your time series exhibit higher values, a larger window size would integrate recent data points, including those elevated values. Consequently, the SMA tends to weight towards the right (later in time), as it accords greater significance to recent data. Conversely, a smaller window size endows the SMA with heightened responsiveness to short-term fluctuations, potentially diminishing its efficacy in capturing long-term trends. For instance, if we visualize a rolling window traversing a time series: as it advances to the right, it integrates more recent data points. Should these recent points exhibit higher values, they would sway the average, prompting the SMA to shift towards the right.

Line 513-530: “It is noteworthy that the correlation coefficient between the time series of SO2 fluxes obtained through assimilation of SO2 column amounts from OMPS and TROPOMI observations, respectively, into the inverse modeling yields a value of 0.55 (Fig. 7a). This modest correlation coefficient primarily arises from OMPS’s constraints in accurately measuring SO2 emissions during both the initial (stage 1) and paroxysmal (stage 2) eruption phases, as noted in sect. 3.1. Therefore, to elucidate the origins of such differences observed in the time series of SO2 fluxes, we investigate the relationship between the spatial resolution of SO2 column distribution and the temporal resolution of the SO2 flux. This inquiry uses the simple moving average (SMA) technique, wherein a window ranging from 2 to 4 hours is applied to the TROPOMI-derived SO2 flux time series. Our observation unveils that with a 4-hour SMA window, the TROPOMI-derived SO2 flux coincides reasonably with the OMPS-derived SO2 flux, proving a correlation coefficient of 0.68 (Fig. 7c). Moreover, upon

assimilating smoothed TROPOMI SO2 column data within a 75-km radius disc (see Fig. 4v), the resultant inverted SO2 flux time series shows a correlation coefficient of 0.69 when compared with the OMPS-derived SO2 flux time series (Fig. 7d). This underscores the advantage of TROPOMI measurements revealing high spatial resolution in ascertaining SO2 flux values at higher temporal resolutions."

19. *L403: Many of the plots shown in Figure S15 show quite an asymmetric distribution, so fitting a Gaussian function does not seem to work well (e.g. panels a, c, d). I would instead suggest fitting an asymmetric function to better capture the difference in the positive and negative values.*

We appreciate your observation. As per your suggestion, we have incorporated the least-median-of-squares (LMS) fit for the residuals, well-known for its robustness against outliers. Consequently, both the main body of the text and Fig. S15 have undergone corresponding revisions.

Line 537-540: "The CTM simulation, initialized with inverted SO2 sources, closely matches to LEO satellite observations, maintaining a least-median-of-squares (LMS) of analysis residuals close to zero throughout the entire eruption period, exhibiting robustness against outliers (Fig. S15). However, the elevated sample mean and standard deviation of residuals, particularly notable on December 16 (Fig. S15d, S15g), highlight the inherent constraints of inverse modeling."

20. *L449-450: In addition to spreading out a plume to obtain lower VCDs, injecting the plume at a higher altitude will have an impact on its location due to wind shear. Is it feasible for winds at lower altitudes to have transported the pre-eruptive plume that you attribute to the pixels in the swath edge, or is this only possible at higher altitudes? If so, then this would suggest that this is not pre-eruptive emissions.*

Your question is appreciated. Indeed, the delineation of pre-eruptive emission itself remains subjective (e.g., Shreve et al. 2022; Smittarello et al. 2022). In this investigation, we denote the #T1 emission (Fig. 1) as pre-eruptive, as approximately 12 hours thereafter, we observe the commencement of the initial stage of eruption. As for the query concerning lower-altitude winds, which are southeasterly during this seasonal period in this geographical region (see Feng et al., 2020), the trajectory of the SO2 plume would veer westward from the source (Fig. S32-ii). Nevertheless, solely the injection of SO2 at higher altitudes would direct it towards the east/northeast quadrant of the source. We have substantiated this phenomenon for emission #T1 by introducing additional supplementary illustrations (Fig. S32). Moreover, this pre-eruptive emission (as per our stipulated definition) is consistently detected at the periphery of the swath by both the LEO instruments, rendering it inadequately constrained by inverse modeling procedure (Fig. S14). Considering the analysis depicted in Fig. S5, the temporal dynamics and injection altitude of this pre-eruptive event may exhibit limitations due to

insufficient constraint. Otherwise, for clarity, we have added further explanations in the main text.

Line 450-452: " Through external calculations using the HYSPLIT trajectory model, we consolidate our inverse modeling retrieval of the injection height (approximately 11 km) of this pre-eruptive emission event (#T1), as it advances towards the northeast quadrant of the domain (Fig. S32)."

21. *L452: When you say "larger wind fields", do you mean higher velocity?*

Yes, we are discussing about the magnitude of wind fields. The sentence has been modified for clarity.

22. *L453-458: Is it possible that using the 7 km VCD product is artificially pushing altitudes higher due to this effect? The retrieved VCD in the 7 km product will be lower than for the plume measured if the reported injection altitudes are similar to the plume altitudes at the time of measurement, so a higher altitude/faster wind may have been selected by the model to account for this.*

In revisiting your fifth major comment, we address the comparison between inverted SO2 flux values and SO2 injection heights through the assimilation of 1 km, 7 km, and 15 km SO2 products from TROPOMI. Notably, while the SO2 mass flux values exhibit variations, the SO2 injection heights remain consistent. As anticipated, varying assumptions regarding the SO2 center of mass altitude yield divergent SO2 column values (Fig. S20-S25), consequently influencing flux amplitudes required to generate corresponding vertical column values within the model prediction. However, the injection heights remain largely unchanged. Thus, concerns regarding artificially elevated altitudes due to the 7 km SO2 product may not hold here. Conversely, the assimilation of a SO2 product considering temporal variations in SO2 center of mass altitude, coupled with accurate SO2 column values, could enhance the precision of SO2 flux values and injection height estimations. Notably, the model's preference for higher altitudes and faster wind patterns to align with observations stems from inadequately constrained SO2 column values, often situated at swath edges or injected at lower altitudes (< 2 km ASL), scenarios absent in CTM simulations. We have added more explanatory sentences for clarity.

Line 600-612: "Before concluding, one final inquiry arises: might the incorporation of the 7 km SO2 product introduce biases in the retrieval of SO2 flux values and injection altitudes due to potential underestimation in instances where the altitude of center of mass of SO2 is below 7 km ASL? To address this query, we conducted assimilation experiments using alternative SO2 products from TROPOMI, namely the 1 km and 15 km SO2 column values (Fig. S33). As expected, variations in assumptions concerning the altitude of the SO2 center of mass give rise to different SO2 column amounts (see Figs. S20–S25), thereby exerting an influence on the requisite flux amplitudes necessary to generate corresponding total vertical column values

within the model simulation. The temporal progression of SO2 flux remains consistent irrespective of the TROPOMI SO2 column product assimilated (at 1, 7, or 15 km). Nevertheless, beyond this temporal correlation, flux values exhibit proportionality, differing by a scaling factor of approximately 3 when transitioning from the 7 km product to the 1 km SO2 column product (refer to Fig. S33). The injection altitudes remain consistent. Thus, the model's inclination towards higher altitudes and faster wind fields to align with observations originates from inadequately constrained SO2 column values, often situated at swath edges, or introduced at low altitudes (<2 km ASL), scenarios not simulated in CTM simulation."

23. *L460: "greatly aids in accurately capturing emission timing" – on this point, if the altitude is incorrect then the emission timing will also be off due to the difference in wind speed. Have you considered the magnitude of this error?*

Thank you for bringing up this query and for your earlier suggestions. Upon assimilating all three SO2 products from TROPOMI into inverse modeling, it has become clear that the assumption on the height of the center of mass of the SO2 vertical profile (1km, 7km, and 15km) has negligible influence on the inverted SO2 flux time series concerning the timing of emissions and injection altitudes. However, as expected, due to fluctuating SO2 column values among the different SO2 products, the emission mass also exhibits variability in the inverse modeling results. Regarding the error in the retrieved altitudes of the SO2 injection profile during the weakly degassing phase of the eruption (sect. 4.2.2), it is relevant to note that our study predominantly focuses on the eruptive phase. This clarification has been provided in section 2.3.

Line 210-214: "At an elevation of 1.3 km ASL, Ambrym's vent stands, prompting a mindful consideration of the constraints inherent in satellite measurements. Consequently, our scope of investigation narrows to emissions surpassing the 2 km ASL threshold, thus characterizing a more precise focus on the eruptive phase of SO2 degassing. Therefore, we do not expect the CTM simulations to generate an SO2 plume to the northwest of the source, as driven by the low-level southeasterly trade winds (see Fig. S32ii)."

24. *L462-464: It is worth noting that the plume height retrievals only work for strong plumes, so may not help for constraining weaker emissions.*

Your observation is correct. We have highlighted this matter in sect. 2.3, as referenced in the preceding response. Moreover, upon analyzing the profiles of the averaging kernels (Fig. S18), the challenge in constraining weak emissions at extremely low altitudes becomes evident. Nevertheless, this underscores the commencement of a new study, wherein we endeavor to integrate such averaging kernel data within the framework of inverse modeling.

25. *L470-471: No mention is made of the trough in SO2 emissions at the peak in lava flow index.*

The inquiry into the intricacies of lava flow dynamics and SO2 degassing has been addressed in response to your 4th major comment.

26. *L485-486: Can you expand on this further? No comment on the physical mechanism of transporting this SO2 from depth to the surface, nor if it came from the lava lake or the lava flow region (or somewhere else?). I would also note that the dyke intrusion lasted ~3 days, so why is the SO_2 emission such a sharp peak? Are there any ground-based observations to support this? Shreve et al. (2019) also note that the total magnitude of SO_2 emissions matches that expected from the volume of the lava flow, so that contributions from the dyke were not significant. I agree that the timing of this peak in SO_2 emission after the lava flow is interesting and may provide insights into the magmatic processes, but this needs further explanation.*

The inquiry into the intricacies of lava flow dynamics and sulfur dioxide degassing has been addressed in response to your fourth significant comment.

27. *Figure 1: in panel bii the #1 arrow is pointing to an empty region near the plume, should this be pointing to the pre-eruptive plume in the swath edge?*

Thank you; the figure has been rectified.

28. *L82: Consider replacing "emissions" with "emission"*

Thank you for the correction.

29. *L212: You refer to points in figures 1 and 2 as "T2" and "O2", for example, but in the figures these are just numbers (e.g. #1, #2). It would be clearer if these were consistent (i.e. use #T1 in figure 1, #O1 in figure 2).*

In the captions of Figs. 1 and 2, we have denoted the significant SO2 emissions or peaks with the symbols "#T" for TROPOMI and "#O" for OMPS. Now, your suggestion is incorporated in these figures.

30. *L299: "SO2" is missing a subscript here.*

Thank you for the correction.

31. *Many figures use non-perceptually uniform colourmaps, which can be misleading or difficult to interpret, especially for colourblind people or when printed in black and*

white (Crameri et al., 2020). I would suggest replacing the colourmaps used throughout with colourblind-friendly versions.

Thank you for your valuable suggestion. We have revised all figures to ensure accessibility for individuals with color vision deficiencies and acknowledged Crameri et al., (2020, 2018).

Line 768-769: "We consistently apply the scientifically calibrated colormaps, which remain perpetually uniform, as described in Crameri et al. (2020); Crameri (2018)."

32. *Satellite figures: The figures of satellite data all have elliptical pixels for the data. Why is this used instead of a continuous grid of rectangular pixels? This would avoid the overlapping in the OMPS data.*

Thank you for your suggestions, but adopting this approach would obscure the advantages afforded by TROPOMI measurements compared to those of OMPS, particularly due to the high spatial resolution inherent in TROPOMI measurements. By presenting elliptically sized pixels generated through satellite altitude, viewing zenith angle, and nadir pixel size, we show the influence of satellite viewing geometry on the pixels at the edge of the swath. This, in turn, emphasizes considerations such as signal-to-noise ratio, the presence of gaps between swaths near the equator, and the detection limit of SO2. However, in response to your recommendation regarding color schemes, we have implemented the scientific color palette endorsed by Fabio Crameri.

33. *Figures S8-11: How have the sub-frames been ordered? They do not appear to be in terms of number, time, altitude or strength of emission and interpreting these plots was difficult. Would it be possible to reorder these, or have I missed the ordering?*

The descriptions of the most influential emissions (termed tracers) have been revised, primarily to enhance the clarity of the color maps and the accompanying descriptions for each sub-frame. These tracers are arranged in descending order (1--28) based on the magnitude of SO2 mass each carries. Presented herein are the first 28 tracers. Their collective sum, depicted in terms of vertical column values, is subsequently juxtaposed with observational data. Notably, within the observational sub-frame (denoted as OBS), only pixels exceeding 0.3 DU are displayed, as these exclusively used in the assimilation process. To provide further elucidation, a sentence has been included in the figure caption.

Reviewer – 2

Major remarks:

1. *The paper seems to lack a scientifically sound method to quantify and compare the inversion result using either TROPOMI or OMPS.*

Thank you for your observation, and we are grateful for the precise and insightful corrections and suggestions provided. We return with further analyses and supplementary material to robustly substantiate our findings derived through inverse modeling. Our endeavor includes a thorough comparison with the TROPOMI and OMPS SO2 column measurements. Concerning inquiries about the inverse modeling framework, it is worth noting that this procedure and its outcomes have been meticulously documented and validated with ground-based DOAS flux observations in scientific publications, such as those by Boichu et al. (2013, 2014, 2015).

2. *It remains often unclear, why they can conclude the strong superiority of TROPOMI data.*

Regarding the preeminence of TROPOMI data within the realm of remote sensing products, especially in comparison to other Low Earth Orbit (LEO) ultraviolet sensors, numerous publications confirm this claim. Noteworthy among these is the work by Theys et al. (2019). We draw attention once more to the satellite observations proximate to the source (Fig. S12, S13, S26-S31) and the distinctive SO2 column values at swath edges (Fig. S14). On our part, the assimilation of such a high-spatial resolution product has enabled the derivation of hourly SO2 flux values, supported by further careful analyses.

In our latest analysis (Fig. 7), we distinctly illustrate how the high spatial resolution of SO2 measurements yields correspondingly high-temporal resolution SO2 flux values, a contrast starkly evident when compared against smoothed (with a disc of 75 km) TROPOMI and OMPS data. Additionally, by combining data from various sources, we illustrate, borrowing from the findings of Shreve et al. (2019), that TROPOMI-derived SO2 flux values align more aptly with seismic activity measured at ground, complemented by the thermal anomalies captured by geostationary Himawari-8/AHI satellite (Fig. 8). This heightened temporal dynamic is notably subdued in the SO2 flux values derived from OMPS data.

3. *The recommended approach would be to use the derived emissions parameters (flux and injection height) in a forward simulation and to develop a metric to quantify to what extend the simulates plumes agree with observations. It is not satisfactory to only juxtapose maps of plumes with HIMAWARI proxy SO2 data.*

We have done the initial phase of our analysis, comprising the model prediction derived from inverted SO2 flux values and injection heights. Subsequently, we perform a visual

juxtaposition of SO2 column values against satellite observations (Figs. 1, 2). Our focus extends to residual analysis, wherein we have refined the residual distribution through LMS fitting (Fig. S15), illustrating a consistent proximity to zero in its peak.

Should the usage of solely the geostationary Himawari-8/AHI SO2 proxy data (Shreve et al. 2019) lack the requisite strength to corroborate our independently computed SO2 flux values via inverse modeling, we have combined ground-measured seismic data (Fig. 8) to consolidate our derived SO2 flux values in this study. Similarly, the determination of SO2 injection heights, derived from our inverse modeling efforts, finds corroboration through external calculations utilizing the HYSPLIT trajectory model. These calculations are initialized by 0.5˚ GDAS reanalysis data sourced from the NOAA's established server, accessed via their web interface (https://www.ready.noaa.gov/hypub-bin/trajtype.pl).

4. *While the authors come to the plausible conclusion that TROPOMI data are much better, it remains unclear how this conclusion is derived especially considering this lack of rigor mentioned above. It should also be emphasized more that they study only one specific episode using their specific modeling framework. I recommend using a more neutral language when describing the results.*

   We have previously addressed these points in response to your 1st and 2nd comments.

5. *The inversion approach should be explained in more detail from a practical perspective. In particular, the robustness of the derived plume heights remains unclear. It is also not clear, what prior information was used and how the ensemble of tracer plumes injected at different height is part of the framework.*

Your concern is noted and appreciated. Our approach deliberately maintains a theoretical framework, facilitating its utility and development by readers—a paramount objective in scholarly articles. To enhance accessibility, we have updated the text, notably refining sect. 2.4 and incorporating pertinent new references elucidating inverse modeling procedures for estimating volcanic SO2 sources. Furthermore, we have strengthened the validation process concerning retrieved SO2 injection heights through the integration of external calculations from the HYSPLIT trajectory model (Fig. 5, S32, S34). In sect. 4.2, we delve deeper into the potential uncertainties inherent in estimating SO2 injection heights, particularly when emissions are minimal, underscoring this as a limitation of our study, which accounts for both satellite measurements and our focused objective of constraining the eruptive phase of the Ambrym eruption.

Line 221-230: “This procedure assimilates total vertical column amounts of SO2 plumes from LEO satellite observations (either TROPOMI or OMPS here), using the well-established data assimilation procedure of source term inversion (Folch and Mingari, 2023). In contrast to traditional methodologies, this procedure obviates the need for a priori assumptions regarding

SO2 emissions, such as flux or injection altitude. Here, inverse modeling entails simulating the dispersion of an ensemble of SO2 (passive) tracers released into the atmosphere using the CHIMERE CTM model. Thereafter, by juxtaposing the modeled dispersion of SO2 vertical columns from the ensemble of tracers with satellite-observed SO2 columns over successive days, the SO2 source (namely, its flux and injection altitude) can be discerned through an inverse problem. This inverse problem aims to find the historical trend of SO2 flux and altitude of injection that minimizes the disparity between observed and modeled spatial and temporal distributions of SO2 vertical columns, often resolved through a least-squares approach using the optimal estimation method."

Line 260-261: "Later on, it shall be noted that the model simulation will be denoted as the product of **G** and $\hat{m}$."

Line 270-275: "The threshold appears from the statistical metrics of the retrieved SO2 source distribution, namely, the mean (μ) and standard deviation (σ). This distribution encapsulates the flux values of the complete ensemble of tracers used in the forward CTM calculation, each tagged with a timestamp and injection altitude."

6. *The paper should provide in a numerical way the derived time series of flux and injection height to allow the scientific community to use the data. If applicable, the result should be compared to the results of other authors.*

The time series of sulfur dioxide (SO2) flux and injection heights, derived through the assimilation of SO2 column measurements from TROPOMI into inverse modeling, are presently accessible within the DATA TERRA repository, an open data repository (https://www.easydata.earth/#/public/home). The DOI is not available now, but we should have it soon.

Furthermore, there is no ground-based DOAS flux data available for Ambrym's 2018 eruption. A rough comparison is made with literature review of past field campaigns in sect. 3.1.1, especially in line 335-360.

7. *The abstract needs to be strongly revised to report in more detail the findings and to reduce the number of the more general or introductory statements.*

While it is typically recommended to make the abstract more accessible by minimizing the use of technical terminology, keeping brevity, and using introductory statements to engage a broader audience, we have taken your suggestion into account. So, we have rewritten the abstract, presented below.

Abstract: "Volcanic eruptions release sulfur dioxide (SO2) gas, affecting air quality, ecosystems, aviation, and potentially climate. To comprehensively assess these atmospheric effects as well as local volcanic hazards, high-temporal-resolution information on SO2 mass flux and injection height is crucial and can be delivered on an hourly basis by inverse modeling.

We show here the strength of assimilating high-spatial-resolution SO2 column measurements from Sentinel-5P/TROPOMI hyperspectral observations, compared to coarser resolution Suomi-NPP/OMPS data, through inverse modeling using the CHIMERE Eulerian chemistry-transport model. As a case study, we investigate the dynamics of the 2018 Ambrym eruption (December 13–18, Vanuatu) starting with the extrusion of an intra-caldera lava flow, coinciding with lava lake draining, followed by the lateral propagation of a voluminous intrusion of magma triggering a submarine eruption. Prior to this eruption marking the end of a decades-long sustained passive degassing associated to lava lake activity, Ambrym held the distinction of top ranking volcanic SO2 emitters in the world. The assimilation of TROPOMI data by inverse modeling proves efficient for accurately characterizing Ambrym SO2 emissions during periods of intense degassing that can be strongly underestimated with OMPS data. We show that this advantage relies on the high spatial resolution of TROPOMI observations which allows for robustly capturing near-source highly concentrated SO2 plumes emitted a few hours before satellite overpass. TROPOMI measurements, also facilitate the detection of volcanic SO2 even at the noisy swath edges, thanks to a superior signal-to-noise ratio and less pixel geometric distortion than OMPS. This distinctive attribute of TROPOMI has enabled the characterization of pre-eruptive SO2 emissions missed by OMPS. Moreover, our inverse modeling procedure effectively discerns, and traces hidden SO2 plumes beneath clouds by assimilating SO2 column data from successive satellite overpasses, hence mitigating satellite retrieval limitations. Furthermore, this approach proves to be less susceptible to interference from ash emissions, as compared to flux estimation derived from near-source observations by the geostationary Himawari-8/AHI sensor. Our study unveils that the 2018 Ambrym eruption discharged approximately 42 ± 16 kt (by OMPS) to 52 ± 13 kt (by TROPOMI) of SO2 during the event. The hourly-resolved SO2 flux time series retrieved by inverse modeling sheds light on the eruption's progression, pinpointing distinct sources of SO2 emissions from either lava flow or shallow magma intrusions. In summary, the assimilation of TROPOMI data into inverse modeling procedures holds substantial promise for advancing our comprehension of magma transport and the environmental repercussions associated with volcanic eruptions."

8. *The paper should get a more precise title*.

   We are sorry, and we shall go ahead with the title we have put forth.

9. *The Figure captions are too long because they interpret the shown results. This should be done in the body text*.

We find this assertion pertains to Figs. 1 and 4 in the main text. However, it does not hold entirely true, as we present the figure captions as stand-alone entities, a practice consistent even in the supplementary materials. Our approach eschews the inclination to present the results within the confines of the figure captions. Undeniably, Figs. 1 and 4 convey a substantial amount of information. This consolidation results in a rather extensive caption. That said, we have indeed revised the caption on Fig. 7.

10. *On the other hand, to little explanation of the meaning of the different lines of the time series is provided. I recommend showing the emission parameter time series as separate plots, not mixed with the maps*.

There is a divergence of opinions at this point. The distinct time series delineated in Figs. 1 and 2 have been thoroughly presented, drawing upon the geostationary Himawari-8/AHI-retrieved SO2 proxy and thermal anomalies as elucidated by Shreve et al. (2019). The partitioning of time series depicting inverted SO2 flux values alongside the SO2 column maps on a daily basis may prove difficult for readers seeking comprehension of these pivotal source terms within the model simulation. In our revised methodology sect. 2.4, dedicated to inverse modeling, we have tried to further elucidate the essence of model prediction.

Minor/Specific remarks:

1. *L10 Please mention, how this was achieved.*

The model simulation now captures the SO2 plume, particularly in regions covered by cloud covers, thanks to the initialization with retrieved SO2 sources via inverse modeling. This achievement is facilitated through the assimilation of successive days of satellite SO2 column data. Notably, revisions have been made to enhance clarity in the figure (Fig. 6) and pertinent text (lines 468-485), as previously suggested by the first reviewer.

2. *L13 Please provide some actual numbers of the emissions here*.

This information has been revised in the latest version of the abstract.

3. *L129 It needs to be properly discussed that the subject comparison with HIMAWARI is the main reference for your study*.

We appreciate your insightful feedback. Indeed, the Himawari-8/AHI reference stands as a pivotal cornerstone in our study. Through the discernment of parallels and contrasts, we draw invaluable insights into our subject matter. Accordingly, we have amended the main text to ensure clarity from the beginning.

Line 114-116: “We compare our findings derived from this inverse modeling procedure with the qualitative estimate of SO2 flux obtained through near-source observations by the geostationary Himawari-8/AHI, as described in Shreve et al. (2019).”

4. *L152 A model top of 200 hPa seems too low to simulate the fate of many volcanic SO2 plumes. Are you sure the plume is always located below that height ?*

The answer is nuanced. Through usage of the IASI SO2 height product, with due acknowledgement to Lieven Clarisse for the data provision, and bearing in mind the inherent limitations and uncertainties, our investigation on the SO2 height product within a 50 km radius of the source (Fig. S19) reveals no instances of SO2 height surpassing 10 km ASL. Nonetheless, on days obscured by cloud cover (December 17-18), which are not depicted herein, selected pixels indicate SO2 heights reaching up to 20 km far away from the source. Subsequent analysis, however, demonstrates that these retrievals are affected by cloud contamination. This discussion has also prompted updates in the main text (lines 205-215), as highlighted by the first reviewer.

Line 768-771: "We extend our thanks to Lieven Clarisse for generously providing us with the IASI SO2 height product. This invaluable contribution greatly facilitated the validation phase of our retrieved SO2 sources through inverse modeling."

5. *L154 Please explain in more detail how the tracer ensemble is used in the inversion framework.*

The ensemble of model parameters, namely the tracers, adheres to the emission method delineated in the works of Boichu et al. (2013, 2014, 2015). Our CTM simulation is structured such that the model emits a tracer—positioned at a specific altitude and time—releasing a fixed quantity of SO2 over the course of a single hour. Subsequently, this emission plume undergoes transport through all pertinent atmospheric physical processes. In this investigation, we employ 1200 passive tracers as model parameters, indicating that the dimension of vector $\boldsymbol{m}$ is 1200x1 (as per Eq. 1 in the main body of text). Furthermore, sect. 2.3 has been revised in accordance with the comments of the first reviewer.

6. *L163 Does the state vector include the emissions, or is the state only the atmospheric concentration.*

Sorry for any confusion incurred. The state vector exclusively encompasses emissions, denoting mass per hour at specific altitudes. We have revised the pertinent sect. 2.4 for clarity and precision.

7. *L170 Please introduce the emissions in your framework here*.

This is done in the revised version.

8. *L160 Please add more information about the time stepping and assimilation window length in the section. What method do you use – a ensemble (KF) or variational approach.*

We regret any confusion that may have arisen. Our assimilation procedure does not adhere to the Kalman filter, nor does it employ the 3D or 4D-Var approach. Sect. 2.4 has been revised accordingly. Our inverse modeling procedure aligns with the least-squares optimal estimation method, as previously expounded upon in lines 82-83. For more clarity, we have included a recent reference (Folch and Mingari, 2023) which refers to this method as the "source term inversion".

9. *L245 This is the first time you mention an indication that TROPOMI agrees better with Himawari than OMPS.*

We have previously implied this assertion in line 210; however, the elucidation of the use of Himawari-8/AHI measurements in the introductory section makes it clearer.

10. *L299 This sensitivity study should be given more attention, perhaps its own section. Was it done only for the outgassing or also for the eruptive events*.

Thank you for directing your attention to this section. It pertains to the period encompassing December 13--18, as previously shown. The correlation analysis encompasses all days within this interval, yet the visual representation of the SO2 column distribution is specifically focused on December 16, the date when the plume achieved its most pronounced manifestation. Following the insightful observations of the first reviewer, we have meticulously revised sect. 3.1.2. Additionally, we have included supplementary figures (Figs. S26-S31) elucidating the application of a 75-kilometer disc smoothing technique to the TROPOMI dataset, followed by its remapping onto the OMPS grid. This approach serves to illuminate the saturation effect clear in OMPS measurements when confronted with elevated SO2 concentrations, notably discernible on December 15 and 16 (for instance, see S28, S29). It is imperative to note that our spatial smoothing methodology is calibrated to preserve the SO2 mass integral within the specified domain.

11. *L311 Discuss the relation to the model resolution.*

Our simulation's spatial resolution closely aligns with TROPOMI measurements (sect. 2.3). By using 0.25˚ archived meteorological data (ERA-5) for initial and boundary conditions, a WRF run might yield minor second-order distinctions between 10x10 km² and 7x7 km². Without conducting another simulation at TROPOMI's spatial resolution, discerning the advantages of conducting a more costly WRF simulation and then driving a CHIMERE simulation at such a high spatial resolution, along with the potential valuable benefits, proves challenging. This inevitably prompts avenues for future research. Moreover, pragmatically, we

apply a radius of 25 km disc spatial smoothing on the CTM simulation prior to the assimilation of the OMPS SO2 columns into inverse modeling due to their coarse spatial resolution than the CTM run.

12. *L322 How do you know that TROPOMI does not overestimate this value ?*

You are right to question this part, particularly now with the emergence of COBRA SO2 retrievals from TROPOMI, recognized for their heightened accuracy (Theys et al., 2021). Nevertheless, within this context, we assert a reduction of approximately 20-fold compared to TROPOMI measurements. Furthermore, with the latest investigation encompassing varied TROPOMI SO2 products, using DOAS algorithm (Theys et al., 2017), namely those at 1 km, 7 km, and 15 km (refer to Fig. S22), our assertion is still valid. This development also paves the way for forthcoming investigations aimed at assimilating COBRA and DOAS SO2 products, with a specific focus on comparative analysis between the two. We have introduced a pertinent statement about this prospect in sect. 5.

Line 732-735: "The newly accessible TROPOMI SO2 column data (Fioletov et al., 2023), derived from the COBRA algorithm (Theys et al. 2021), presents a significant advancement in accuracy and reliability over prior methodologies. With heightened sensitivity in detecting even minor emission sources, this development marks a promising avenue for assimilation into transport models aimed at characterizing volcanic SO2 sources."

13. *L333 Is this bimodal injection height something that has been observed before - or is it an artefact of the assimilation, which does not get enough information about the vertical distribution of the plume*.

We adopt a bimodal terminology to explain our findings, as illustrated in Fig. 3, wherein both the SO2 sources show two prominent altitude bands of emissions, notably at 5km and approximately 9-10 km. The validation of these distinct injection heights is depicted in Fig. 5, where corroborative evidence is drawn from external sources including geostationary Himawari-8/AHI SO2 RGB images (Fig. 5-i--vi). These images vividly delineate a bifurcation in the SO2 plume, suggesting the possibility of dual (bimodal) altitude injections. Additionally, trajectory calculations conducted via the HYSPLIT model employing 0.5˚ GDAS reanalysis data further support this assertion. The frequency plot of trajectories (Fig. 5vii--ix) unequivocally shows a divergence contingent upon the altitude of initialization.

Concerning the data on the vertical distribution of the plume, no specific profile is presumed at any stage of the eruption in the inverse modeling procedure. A total of 1200 tracers (the state vector, or model parameters) are emitted on an hourly basis from December 13--18, spanning altitudes from 2 to 11 km, with a vertical resolution of 1 km (Fig. S3).

14. *L336 Please provide more details. HYSPLIT has not been introduced before to be part of the study. Did you run the model yourself?*

The inquiry about our use of the HYSPLIT trajectory model was not entirely self-prompted. This model, integral to atmospheric studies, stands open to all individuals for execution via the NOAA server (https://www.ready.noaa.gov/hypub-bin/trajtype.pl). Additionally, we have directed attention to the comprehensive elucidation of the HYSPLIT model by Stein et al. (2015). Nonetheless, we include an additional sentence here to ensure further clarity.

Line 430-435: "Furthermore, through external Lagrangian model calculations (HYSPLIT trajectory model (Stein et al., 2015)), driven by 0.5-degree GDAS reanalysis, it is demonstrated that the release of SO2 at 00:00 UT on December 15 at an altitude of 5 km ASL (Fig. 5-vii) effectively replicates a specific branch of the plume, characterized by its north-northeast movement. This trajectory aligns closely with observations made by the geostationary Himawari-8/AHI (Fig. 5iv–5vi)."

15. *L343 If HIMAWARI is you reference, why is the better agreement between OMPS and HIMAWARI not a good result for OMPS?*

Your observation is well-placed. The crux of our argument about the retrieval of pre-eruptive SO2 emissions rests upon the assimilation of TROPOMI SO2 measurements. This retrieval is made possible by TROPOMI's consistent capture of these SO2 parcels at the swath edge, notably between December 14-16 (Fig. S14). A capability absents in OMPS measurements. To support this assertion, we have confirmed our findings through external calculations using the HYSPLIT trajectory model (Fig. S32).

Concurrently, it is pertinent to note the indispensable role played by Himawari-8/AHI measurements in our study as a reference point. The absence of discernible activity in the Himawari-8/AHI SO2 RGB image during this time undoubtedly raises queries. Regrettably, our case study lacks more observations that could substantiate our retrieval.

Perhaps the question/confusion could be resolved with the availability of the TROPOMI SO2 height product (Hedelt et al., 2019), or more advanced COBRA TROPOMI product (Theys et al., 2022). Regrettably, such data is still unavailable for the specified period under investigation.

16. *L375 This is a very controversial statement that needs more backing up with evidence. Given the high variability of the plumes, using the observations from the previous day to substitute cloud-contaminated observations seems risky.*

Hence, it is imperative to acknowledge the constraints inherent in such procedures. If the SO2 plume persists under the clouds over several consecutive days, our ability to pinpoint the SO2 sources diminishes significantly. However, through the method of sequential assimilation of daily SO2 column values—many of which are expected to be completely free

from clouds—we can successfully find the SO2 source terms. Our inverse modeling results are here confirmed using observations from the geostationary Himawari-8 satellite, alongside corroborating calculations from the HYSPLIT model. Additionally, the high spatial resolution of SO2 column values provided by TROPOMI enables us to confirm the veracity of our model predictions, thereby confirming the retrieved SO2 sources. Addressing the comments of the first reviewer, we have further refined our representation (see Fig. 6), thereby enhancing its clarity substantially.

17. L416 Why initialized? The emissions should be injected during the model run.

Indeed, we are speaking of the same phenomenon. The term "model initialization" denotes the act of instantiating the emission, retrieved (by inverse modeling) at a specific time and altitude, bearing a designated mass of SO2. This nomenclature aligns with the standard practice of employing initial and boundary conditions to execute an atmospheric model.

18. L487 This conclusion is not clear in this context.

Sect. 4.3 has undergone a comprehensive rewrite, incorporating pertinent references, prompted by queries raised by the first reviewer. Nonetheless, through the new analysis of assimilating smoothed TROPOMI data by a disc of radius 75-kilometer, we unequivocally demonstrate that measurements of high spatial resolution facilitate the retrieval of SO2 source terms with heightened temporal resolution via inverse modeling (see Fig. 7a).

19. L495 Provide more reasons.

Please refer to the revised segment 4.3 (response to 4th major comment by the first reviewer).

20. L496 It is not clear why a high spatial resolution is helpful for capturing short - lived (short?) emissions. The overpass intervals is the same for TROPOMI and OMPS.

Please refer to the updated Fig. 7a for further clarification. It is now clear here. With our recent analysis, as previously noted, we have assimilated a set of SO2 column data, specifically (smoothed) TROPOMI data. Subsequently, in Fig. 7a, we illustrate the consequence of applying spatial smoothing with a 75-kilometer radius, resulting in a discernible loss of spatial variability in the distribution of SO2 column values (Fig. 4). Furthermore, this spatial smoothing also compromises the temporal resolution of SO2 flux values (as depicted in Fig. 7a). This observation underscores the notion that greater spatial variability keeps a form of memory, easing the retrieval of high temporal resolution SO2 sources through inverse modeling procedure.

Notably, the synchronous overpass time of both the LEO sensors confers a significant advantage to this study, enabling a robust comparison of SO2 measurements and enhancing the efficacy of the inverse modeling procedure.

21. L501 That is a very general statement, please provide evidence from the study.

The robust reaffirmation of the capacity to reconstruct SO2 source terms through inverse modeling, achieved by assimilating successive days of satellite derived SO2 column data, highlights the notable advancement shown in the refined depiction of Fig. 6 and the clarified text in Sect. 3.2. This enhancement offers a compelling demonstration of the model's capability to discern SO2 plume locations undetected by satellite observations because of the presence of meteorological clouds.

22. L503 Again, I am not convinced that has been proven universally in that paper. Please explain if you mean the approach to use yesterday's observations to gap fill.

The clarification of this confusion has been addressed; a concern initially raised by the first reviewer. For further explanation, we direct you to our response concerning the 11th minor point posited by said reviewer.

23. L508 Compare that to values from the literature.

If our interpretation is correct, your concerns pertain to the aggregate mass of SO2 retrieved by inverse modeling, referenced in line 506 rather than line 508. Please acknowledge that this mass has already been cited and compared in lines 267-268 of the submitted draft.

24. Figure 1,2 Consider putting the time series in a separate picture. Please explain in more detail all 4 lines in the time series plot.

The disjunction between the time series of SO2 flux and model predictions presents a challenge to discerning the significance of inverted-SO2 flux values in the predictive model—a point already addressed in your major commentary. Apart from this, we remain uncertain as to the specific other information you seek about the four time series illustrated in Figs. 1 and 2. We have undertaken a revision of sect.2.4, aiming to provide clarity without unduly extending the length of the figure captions. This revision should prove illuminating.

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