July 1, 2014

Estimating the volcanic emission rate and atmospheric lifetime of SO_2 from space: a case study for Kīlauea volcano, Hawai'i by S. Beirle et al.

Reply to anonymous reviewer #3

We thank Reviewer #3 for his constructive comments and suggestions. Below we reply to them one-by-one.
Green: Reviewer comment.
Black: Author's reply.
Red: Modified text in manuscript.

Reviewer comment: This paper introduces a new technique that allows volcanic SO2 emission rates and SO2 atmospheric lifetimes to be simultaneously derived from GOME-2 satellite imagery and modeled wind fields. The authors present results from a case study at Kilauea volcano, where they detect higher emission rates than ground-based methods. This paper is well written and provides a new method for determining SO2 emission rate and SO2 loss simultaneously. The interesting finding of significantly higher SO2 emission rates determined from the GOME-2 satellite data relative to ground-based measurements is intriguing, but in my opinion requires further investigation (or elaboration) to determine if the estimated GOME-2 emission rates are in fact accurate. This observation is inconsistent with several previous satellite validation studies (e.g. Bani, JVGR, 2012; Lopez, JVGR, 2012; McCormick, G-Cubed, 2012), which have found satellite SO2 emission rates to be lower than ground-based measurements. The authors propose that underestimation of ground-based measurements due to previously unaccounted for radiation transfer effects (Kern et al., 2013) can account for these differences; however I would expect that these effects may also have influenced the ground-based measurements used in the other satellite validation studies?

Authors' reply: The referee is right in pointing out the apparent inconsistency. There are several reasons for the observed discrepancies, which will be mentioned shortly here and in more detail in the revised manuscript. We have extended the description of the GOME-2 SO_2 retrieval and the discussion of uncertainties. We thereby substantiate that Kīlauea emission rates (despite the relatively large uncertainties of about 40%) are significantly higher than reported from ground-based measurements, at least in 2008.
As we have now clarified in the manuscript, the presented Kīlauea dataset is unique in that the ground-based measurements were typically made less than 1 km from an extremely prodigiously degassing vent. Thus, the encountered SO_2 column densities ($>10^{19}$ molec/cm^2) and aerosol optical depths (>10, values from Kern et al, 2012) were significantly higher than is usually the case. Therefore, in this particular case, SO_2 emission rates derived from ground-based traverses using standard retrieval schemes significantly underestimate the true emissions (Kern et al 2012). Satellite instruments, which are able to measure the plume farther downwind where it is more dilute, have a more favorable measurement geometry.
The reviewer refers to several studies reporting on satellite SO_2 emission rates to be lower than ground-based measurements. We relate part of the reported discrepancies to the a-priori assumptions on the plume profile in the satellite retrieval, which can lead to emission rate estimates that are biased low. For instance, Bani et al. (2012) assume a constant plume height of 3 km, while actual plume heights for Yasur volcano were below 1 km. This has a large effect on AMFs of about one order of magnitude (see the comparison of OMI products for TRL and PBL profiles shown in Lopez et al., 2013, Fig. 5).

Reviewer comment: Additionally, the author's reference to emission rates in units of mass (instead of mass/time) (possibly a typo?), compounded by the large discrepancy with ground-based measurements, and the absence of further validation data, makes it unclear whether their emission rates are in fact comparable to ground-based measurements.

Authors' reply: We determine emission rates in units of mass per time (kilotons per day), which are directly comparable to ground-based emission rate estimates. For 2008, we also compared total emissions, i.e. emission rates integrated over time, resulting in units of mass, as this was a quantity provided by Eguchi et al. (2011). We have corrected "emission rates" to "integrated emissions" in this context.

Reviewer comment: I think that the following changes could significantly improve the impact and utility of this paper: (1) The authors should include a more detailed description of what they are reporting as emission rates and how the comparison (ground-based) measurements were collected (e.g. what downwind distance, what windspeed?) to convince the readers that a fair and direct comparison was made;

Authors' reply: We have clarified the description of the determination of emission rates from the satellite measurements. In addition, we have now included a more detailed description of the measurement location used for the ground-based FLYSPEC traverses. We also mention the source of the wind speed used for deriving the FLYSPEC emission rates (meteorology stations at HVO and Hōlei Pali).

Reviewer comment: (2) The authors should compare their emission rate method and findings to the numerous previous studies in the literature and explain why their observations are different;

Authors' reply: We have expanded the discussion with a section dealing with other methods of emission rate estimates from satellite observations in literature, and clarify how far our approach is different, and why this is necessary/meaningful for the specific conditions for Kīlauea. We relate the "difference" (i.e. the discrepancy between satellite and ground-based estimates) to non-linear radiative transfer effects, which are particularly strong for emissions from Kīlauea summit as a consequence of the short distance of the FLYSPEC measurements to the vent.

Reviewer comment: and (3) The authors should explain in greater detail, including additional supporting data if possible (e.g. a different satellite sensor or chemical model), why the observed discrepancy between ground and GOME-2-based SO2 emission rates is real. Upon addressing these main points, I would recommend this paper for publication in Atmospheric Chemistry and Physics.

Authors' reply: We have expanded the discussions accordingly and explain in greater detail, why we consider the emission rate estimates from satellite measurements to be real, while the ground-based measurements probably underestimate the SO_2 concentration, for reasons mentioned in our reply to the referee's first comment. In order to check the consistency of our GOME-2 SO_2 data product to other satellite data products, we also investigated the operational NASA OMI SO_2 Level 2 product for August 2008. Therefore, the results from the Planet Boundary Layer product (PBL-SO2, assuming a mean altitude of 0.9 km) and the Lower Tropospheric Column product (TRL-SO2, assuming a mean altitude of 2.5 km) were interpolated to an altitude of 2 km. The resulting SO_2 fluxes as function of time are very similar compared to our results from GOME-2:

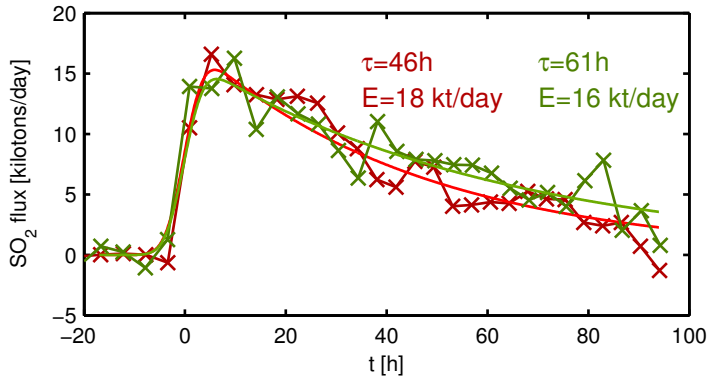

Fig. R1: SO_2 flux as function of time (crosses) and the fitted model function (smooth line) for GOME-2 (red) and OMI (green) in August 2008.

The derived lifetime based on OMI is longer by 33%, as in the OMI monthly mean the downwind are slightly higher than for GOME-2. The estimated emission rates, however, agree within 11%. The deviations are thus smaller than the total error of about 40% estimated in the revised manuscript.
We conclude that our GOME-2 retrieval is in good agreement with the operational OMI product, derived

by a completely independent retrieval algorithm, and that monthly means are meaningful for this approach, i.e. sampling effects are negligible.

Reviewer comment: Technical Revisions: Intro, line 21: Suggestion to specify that this SO2 lifetime only considers homogeneous reactions.

Authors' reply: We consider the sentence sufficiently specific, as "respective" directly refers to the previous sentence ("In the gas phase").

Reviewer comment: Page 28697, line 6: Suggestion to start paragraph with "Kilauea" and use "it" later in the sentence.

Authors' reply: We changed the sentence accordingly.

Reviewer comment: P 28697, line 16: Suggestion to specify "satellite sensors" instead of "they"

Authors' reply: The respective paragraph was largely revised.

Reviewer comment: P 28697, line 21: Suggestion to remove "see" and put "e.g. ..." in parentheses.

Authors' reply: Done.

Reviewer comment: P 28697, line 25: Please spell out ECMWF for its first usage in main text.

Authors' reply: Done.

Reviewer comment: P28698, line 5: Please spell out local time (LT) for its first usage.

Authors' reply: Done.

Reviewer comment: P 28698, lines 7-8: Suggest rewording to: SO2 concentrations integrated along the mean light path, referred to as SO2 Slant Column Densities (SCDs), are derived from...

Authors' reply: Done.

Reviewer comment: P 28698, lines 13: Suggest replacing "aerosol scenarios" with "aerosol optical depths (AOD)"

Authors' reply: Done.

Reviewer comment: P 28698, lines 20-21: Please describe the gridding method ? is this an interpolation? If so, please explain the methods used and/or provide a reference. How do the pixel size and grid size compare to the plume size? Do these size differences influence your results?

Authors' reply: We grid the individual GOME-2 ground pixels, which have an extent of 40x80 km^2, onto a regular 0.1°grid (Each grid pixel covered by a satellite pixel is assigned with the respective VCD). I.e., the grid resolution is much finer than that of the original measurement. In order to illustrate the GOME-2 ground pixel size and allow a comparison with the extent of the SO_2 plume, we have added the footprint of a typical GOME-2 ground pixel to the legend of Fig. 2:

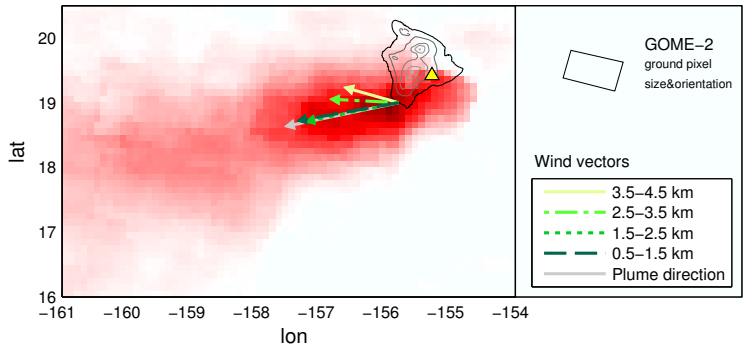

Fig. 2 revised

The cross-track extent of a GOME-2 ground pixel of 80 km broadens the derived SO_2 line densities and the SO_2 flux as function of time. This effect is appropriately accounted for by convolving the model function $F(t)$ with a Gaussian with $\sigma = 80$ km, divided by the wind speed u (see section 2.4).

Reviewer comment: P 28698, line 25: Suggestion to add "-in" after zoomed, e.g. "zoomed-in"

Authors' reply: Done.

Reviewer comment: P 28699, lines 2-5: Please provide references

Authors' reply: We have added references to Beirle et al., 2009, and Leitao et al., 2010.

Reviewer comment: P 28699, lines 7-8: This sentence reads awkwardly, please reword. Possible sugges- tion: Aerosols are formed within the volcanic plume by conversion of SO2 to H2SO4 during formation of, or uptake in, aqueous droplets.

Authors' reply: Done.

Reviewer comment: P 28699, lines 13-20: This section is confusing, can the authors please clarify why the "real" AOD's are not used in the initial evaluation?

Authors' reply: The direct consideration of the "real" AOD in the initial evaluation would complicate and slow down the retrieval, as each satellite observation would require individual RTM calculations. But since the effect of aerosol AOD on AMFs (in the considered range of AOD) is monotonous and almost linear, we decided to correct for AOD effects by
1. calculating three sets of VCDs for different a-priori AOD, and
2. interpolating the resulting VCDs according to the actual AOD.
We have clarified the section accordingly.

Reviewer comment: P 28700, lines 15-16: This sentence is confusing, please reword. Possible suggestion: We determine the mean plume direction using the slope of a weighted linear fit equation applied to the SO2 VCD for lat/lon coordinates of all grid pixels with a VCD above 3x1016 molec/cm2.

Authors' reply: As both reviewers point out that the description of the weighted linear fit was difficult to follow, and in order to avoid a potential bias due to the high VCDs at the volcano over the Island, we have modified our procedure. In the revised version, we apply a simple linear fit without any weights, reducing the impact of the Island pixels. This procedure results in essentially the same plume directions, and simplifies the

description to
We determine the mean plume direction by fitting a line to the lat/lon coordinates of those grid pixels exceeding 3×10^{16} molec cm^{-2}.
The ECMWF wind direction is averaged for the respective area covered by the SO_2 plume for 4 different altitude levels. The choice of the threshold defining the plume extent has only small impact on the results.

Reviewer comment: P28700, lines 20-23: Please refer the reader to figure 2 here.

Authors' reply: Done.

Reviewer comment: P28701, line 5: Suggestion to add "sensor" after satellite and "lower altitude" before emissions: e.g. Therefore, the satellite sensor will be less sensitive to these lower altitude emissions than those originating from the summit vent.

Authors' reply: Done.

Reviewer comment: P28701, line 7-9: Please specify what plume altitude was used for the other years between 2007-2012.

Authors' reply: We do not explicitely specify the plume altitude for the other years, but by applying the relation shown in Fig. 6, it is implicitely assumed that plume characteristics are same. As this is probably not appropriate for 2007 (see above), we now explicitly state that this simple estimate is very likely biased low, and we mark the values for 2007 with a light shading in Fig.7 for clarity.

Reviewer comment: P 28701, lines 11-13: Please provide numbered values to go along with this statement (i.e. specify the ranges determined by Elias and Sutton and Eguich et al.)

Authors' reply: We extended the respective paragraph and added the altitude information provided by Elias and Sutton and Eguchi et al.

Reviewer comment: P 28700-28701, section 2.3: Do you have any constraints on plume thickness (maybe from CALIOP data)? Is the bulk of the plume <= 1 km thick?

Authors' reply: The CALIOP measurements shown in Eguchi et al. indicate a rather narrow plume.
We have calculated AMFs for standard boxes of 1 km thickness, and investigated the impact of mean plume altitude on our results. Compared to this large effect, the actual plume thickness has only small impact on mean AMFs.

Reviewer comment: P 28701, line 18: Suggestion to delete "stable meteorological conditions, i.e." (unnecessary text)

Authors' reply: Done.

Reviewer comment: P 28701, lines 20-23: I found this sentence describing line densities to be a bit confus- ing. Please clarify what the width of each line (for a LD) is (1 degree grid or 1 pixel? provide units). Please clarify that these LDs represent a cross-section of the plume in a direction perpendicular to plume direction.

Authors' reply: Our analysis starts with monthly mean maps of SO_2 VCDs, i.e. SO_2 per area with unit molecules/cm^2. The chosen grid size (0.1°) determines the level of detail in spatial distribution, but does not mean the width of each column. Next, we integrate the column densities in latitudinal direction from 10 to 25°. By this integration, one spatial dimension is cancelled, such that the resulting quantity is SO_2 per length (molecules/cm), therefore we call it a line density, which is a function of the remaining spatial variable (longitude). Again, the grid size determines the level of detail, but does not mean the width of each line. It is important to note that the LD does not represent a cross-section, but is determined by integration. Thus,

dilution effects are mostly accounted for.
The procedure should be clearer in the revised manuscript (section 2.4 and discussion in 4.2.3 (b)).

Reviewer comment: P 28701/28702: Suggestion to combine sections 2.4 and 2.5 as they are quite related.

Authors' reply: We follow the reviewers proposal and label the new section 2.4 as Determining SO_2 emission rates and lifetimes.

Reviewer comment: P 28702, lines 14 - 18: This text is redundant with section 2.4 and could be deleted if not necessary.

Authors' reply: We agree that the text is redundant to some extent, but the respective paragraph is intended to provide those readers familiar with Beirle et al. (2011) with a compact explanation of why and how the original method was modified.

Reviewer comment: P 28703, lines 1-3: I'm skeptical that plume dilution is not contributing to the observed down-wind SO2 depletion. In Figure 2 it can be seen that as the plume travels down- wind it will spread laterally, such that the SO2 VCD will decrease while the plume width increases. This will likely result in some pixels along the edges of the plume having insufficient SO2 to be above detection limit and will likely contribute to some of the ob- served downwind SO2 depletion (e.g. Lopez et al., JVGR, 2013). I would suggest that the authors soften their wording and clarify that they are assuming that the temporal changes are due primarily to chemical reactions and that they are considering dilution to be negligible.

Authors' reply: The key to our approach is that we explicitly consider the effect of lateral plume broadening by the integration of column densities over the complete latitude range of 10-25°. Only if part of the SO_2 plume "leaves" this latitude range, the effect mentioned by the reviewer occurs.
In fact we see this effect if we narrow the latitudinal range down to 15-20°(see table 2 in the revised manuscript): In this case, the SO_2 "loss" by dilution is interpreted as a lower lifetime (17%). On the other hand, a broadening of the latitude range up to 5-30°does not increase the fitted lifetime any further. Thus we conclude that the effect of "loss" by dilution out of the chosen latitude range is negligible.

Reviewer comment: P 28703, lines 10-14: Suggestion to move this text to the figure caption.

Authors' reply: We prefer to keep this detailed information in the plain text, but slightly modified the text and added the months to the figure caption.

Reviewer comment: P 28703, lines 2-6: Please clarify if the previously determined SO2 lifetimes in the literature are also for studies involving volcanic plumes (which may have a higher proportion of surfaces/particles on which to react) or if these studies are referring to industrial emissions? If the latter, I suggest comparing your SO2 lifetimes to those from other volcanic studies (e.g. Oppenheimer, GRL, 1998; Bluth and Carn, IJRS, 2008; Theys, ACP, 2013; McCormick, G-Cubed, 2013; McCormick, JGR, 2014, etc.)

Authors' reply: We assume that the reviewer is referring to P 28704. We have extended the respective paragraph accordingly and included additional references.

Reviewer comment: P 28704, lines 11-13: I think this sentence is a bit misleading as it implies that the authors have taken repeated measurements of the same SO2 within the plume at different down-wind distances. They have not done this, but rather have evaluated SO2 VCDs over time by averaging a series of snap-shots collected every 1.5 days over a month time period, and assuming that any temporal variability in the SO2 emissions (over the analysis area) is cancelled out. I would suggest either deleting the last part of this sentence (i.e. delete: and the actual evolution of SO2 column densities is quantified over time as opposed to simply taking a single snapshot), or softening the wording.

Authors' reply: We agree that the sentence is misleading and reworded the sentence, clarifying that the

"measurement of the plume" from GOME-2 represents a monthly mean composite.

Reviewer comment: P 28704, lines 16-20 and throughout results: It is not clear what "emission rates" the authors are referring to. Are they referring to the max emission rate detected (near t = 0 hrs) during the 1-month average near the plume source? If so, please state this. Or, are they referring to an emission rate calculated from a line density a certain down-wind distance? If so, please specify what distance. Alternatively, if they are referring to their time integrated SO2 emission rates and are including the entire set of emission rates at various plume ages this would represent an SO2 mass. In this latter case, that would not be a true emission rate (e.g. Kg/s vs. Kg/s x s) and the authors would be inflating their results. Also, there are many other papers that summarize methods for calculating SO_2 emission rates from satellite data (e.g. Carn et al., GSL Special Publication, 2013; Theys et al., ACP, 2013). Can the authors comment on the advantages/disadvantages of this technique over the other methods in the literature?

Authors' reply: In section 2.4, the description is given (and extended in the revised paper) on how the downwind flux of SO_2 as function of time is derived from the monthly mean column densities. The complete flux as function of time over the time interval -20 to 100 hours (red data in Fig. 4) is used for the determination of emission rates and lifetimes (rather than just taking the maximum or data at one selected distance). The least-squares fit of the model function $F(t)$ to the observed flux yields then the SO_2 emission rate E in units of kt/day and the first-order time constant τ simultaneously.
In principle, the emission rate could also be derived from the maximum flux close to the source. However, this is complicated by the comparably large ground pixel size of GOME-2, which leads to a smearing out of maxima at the source. This effect is accounted for in our method by convolving $F(t)$ with a Gaussian function, representing GOME-2 resolution. Consequently, the fitted emission rates (Fig. 5) are higher than the maximum SO_2 fluxes (Fig. 4) by a factor of about 1.2.
We added a discussion of the proposed method vs. other approaches for emission rate and lifetime estimates given in literature (as reviewed by Carn et al., 2013 and Theys et al., 2013) in a new section (section 4.1).

Reviewer comment: P 28704, line 21-22: Again the authors need to specify what this emission rate is? Is this the mean SO2 emission rate calculated over time and space (which I would argue is actually an SO2 mass)? Or the maximum emission rate calculated at a specific line-density location (a true emission rate), and if so, what location? I would expect that the average SO2 VCD over the same grid would have a positive correlation as the SO2 mass (scaled by the factor wind-speed), but this correlation does not represent true emission rate, and therefore does not seem to be a significant finding (not worth a figure?). If the authors did calculate a true emission rate, than this correlation would have greater meaning and could justify the use of a figure.

Authors' reply: The displayed emission rates are the fitted values for E (Eq. 1), as described in more detail above and in the revised manuscript.
We expect a high correlation between E and the mean VCD (which should be proportional to $E \times \tau$), but beyond correlation, we also use the fitted slope of Fig. 6 for a rough estimate of emission rates for other years than 2008.

Reviewer comment: P 28704, line 21-22: Please clarify over what area the SO2 VCDs are averaged. This information is included in the figure caption but not in the text.

Authors' reply: We add this information to the text as well.

Reviewer comment: P 28704, lines 28-29: Please add that this also implies that plume speed doesn't change with time.

Authors' reply: We have changed the respective sentence to
Note that this assumption of a linear relation between emission rates and mean column densities implies that conditions like the SO_2 AMFs and lifetime or wind speeds are comparable for the different months.

Reviewer comment: P 28705, line 1-2: Please clarify how effective SO2 lifetime is derived from Emission rate vs SO2 VCD?

Authors' reply: Spatial integration of the mean VCD over 17-20°N, 155-160°W results in a total amount of SO_2, which equals $E \times \tau$ according to mass balance. This has been clarified in the manuscript.

Reviewer comment: P 28705, lines 17-19. This is confusing as written. It may help the readability if the authors specify first that the unusual westerly winds pushed the plume into what had been designated as the "background" area?

Authors' reply: We revised and clarified the paragraph accordingly, and shifted this detail into a footnote.

Reviewer comment: P 28705, line 22: suggestion to replace "made" with "assumed" or "used" to clarify that these were not results but rather selected values.

Authors' reply: As suggested, we replaced "made" with "used".

Reviewer comment: P 28705, line 25: suggestion to replace "Alternatively" with "Additionally"

Authors' reply: The respective section was largely revised.

Reviewer comment: P 28706, lines 3-5: Awkward sentence, please reword.

Authors' reply: We changed the sentence to
This is probably resulting from cloud heights scattering around the SO_2 plume height: apparently, the effects of shielding (clouds above SO_2 plume) vs. multiple scattering (clouds at or under plume) cancel out, at least partly.

Reviewer comment: P 28706, lines 13-15: I would again argue that dilution is a factor (see above elaboration). Please consider rewording.

Authors' reply: As a broadening of the latitude range does not affect the analysis significantly, dilution effects are in fact negligible.

Reviewer comment: P 28706, line 17: It may help if the authors clarify that -20 h represents data from up-wind pixels.

Authors' reply: Done.

Reviewer comment: P 28707, line 2: Suggestion to remove this comma to improve readability

Authors' reply: Done.

Reviewer comment: P 28707, line 5-6: This sentence was hard for me to visualize. I think it will help read- ability if the authors would write-out the effects (i.e. instrument sensitivity and horizontal wind speed?) and also write out explicitly what causes the increase in emission rate estimates (e.g. a lower plume altitude result in both higher SO2 VCDs and higher hor- izontal wind speeds, both of which will contribute to higher estimated emission rates.)

Authors' reply: The respective section is largely revised. In particular we have added a discussion of the uncertainties of a-priori on VCDs as well. I.e., the two effects of a-priori plume height (on AMF and on wind) are now clearly separated.

Reviewer comment: P 28707, lines 7-10: Could the authors please summarize briefly the difference in results from their conference presentation?

Authors' reply: The conference presentation contains an earlier version of this study, with only slight differences in the applied methods and settings. Main difference was the a-priori plume height, which was estimated to be 2.5 km at that time. Consequently, the reported emission rates in Beirle et al., 2012, are lower by about 25%, and lifetimes are higher by about 30% (compare table 1). This is clarified in the revised manuscript.

Reviewer comment: P 28708, line 2: Suggestion to replace "artifacts" with "biases"

Authors' reply: Done.

Reviewer comment: P 28710, lines 20-22: Please state the down-wind distance where Elias and Sutton collected their emission rate measurements.

Authors' reply: Done. These were collected between 0.3 and 1.3 km from the summit vent, and at 9 km from Pu'u 'Ō'ō.

Reviewer comment: P 28710, lines 1-3: Please see concerns above. This result is very surprising to me based on previous satellite/ground-based SO2 comparisons. The authors must specify how exactly they are calculating emission rates and that their methods are in fact comparable to the ground-based methods.

Authors' reply: We hope that this point becomes more clear now that we show how the Kīlauea dataset is unique.

Reviewer comment: P 28710 lines 10-13: Please elaborate on why these RT effects for high SO2 columns will not affect the satellite DOAS retrievals as significantly as ground-based measurements? Are the effects similar, but resulting (inflated) SO2 VCD values are then offset (reduced) by other factors (e.g. dilution?).

Authors' reply: The satellite measurements are generally affected by the same physical processes, but due to the measurement geometry and in particular the ability to make measurements tens to hundreds of kilometers downwind where the plume optical depth is orders of magnitude lower, the satellite measurements are likely more reliable here. We have described this in more detail in the text.

Reviewer comment: P 28710, lines 16-17: These are mass values, not emission rate values. Please clarify how masses were calculated from emission rates (are they cumulative measurements? Were interpolations made between measurements? If so, how?)

Authors' reply: Line 16 should read "emissions" instead of "emission rates"; we have changed the manuscript accordingly. Total emissions result from annual integrated emission rates. The numbers are taken from the cited reference (Elias and Sutton) and "were calculated by summing daily emission rates generated using a non-parametric digital filter".

Reviewer comment: P 28710, lines 19-25: It may be worth mentioning previous ground and satellite based comparisons of volcanic SO_2 and how your findings compare to these previous studies

Authors' reply: We have added a discussion of other approaches for estimating emission rates from satellite measurements in the new section 4.1. However, we keep this case study focused on the situation at Kīlauea, which is particular for both ground measurements (close to the vent, high OD) and the interpretation of satellite measurements (steady trade winds, well defined plume).

Reviewer comment: P 28710, lines 16-27: Did you use a lower plume altitude for these measurements? Or did you assume the same plume altitude (1.5-2.5 km)?

Authors' reply: For other years than 2008, no direct retrieval was applied, i.e. no plume altitude was assumed explicitly. Implicetly, however, the same conditions as for 2008 are assumed by making use of the correlation between emissions and mean column densities as shown in Fig. 6. We have clarified this in the revised manuscript and mark the estimates for 2007 with a different shading, as this assumption is likely not quite appropriate for emissions from the East rift.

Reviewer comment: P 28711, line 2: Please clarify that the more dilute plume is also assumed to be more transparent (optically thinner).

Authors' reply: The respective section was revised considerably, and the aspect should be clarified now.

Reviewer comment: P 28711, lines 7-12: This is awkward as written. Suggestion to reword: For this time period, the discrepancy between satellite and ground-based emission rates instead appears to be caused by the lower plume altitude in 2007, which affects the satellite AMF. As mentioned before, the difference in altitude between the Kilauea summit and East Rift emission plumes leads to a different sensitivity of the satellite instrument towards SO2 emitted at each of the two locations.

Authors' reply: We thank the reviewer for this suggestion, and revised the respective paragraph accordingly.

Reviewer comment: P 28711, lines 25-26: Have the authors compared their results to a model? If so, please share their results. Since the comparison results with ground-based measurements presented here were quite different it would be helpful to have another method validate the author's technique. Comparison with model results or another satellite sensor would be insightful. If the authors have not yet done this, they could be mentioned this as future work to validate their method.

Authors' reply: We consider the comparison to a chemical model to be out of the scope of this study.
In order to assess the reliability of the satellite SO_2 evaluation, we have compared our GOME-2 SO_2 VCD to the operational OMI product for August 2008 and found good agreement (see Figure R1).