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Atmospheric Chemistry and Physics Discussions

Interactive comment on "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.

Anonymous Referee #3

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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? 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. 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; (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; 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.

Technical Revisions:

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

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

P 28697, line 16: Suggestion to specify “satellite sensors” instead of “they”

P 28697, line 21: Suggestion to remove “see” and put “e.g. . . .” in parentheses.

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

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

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. . .

P 28698, lines 13: Suggest replacing “aerosol scenarios” with “aerosol optical depths (AOD)”

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?

P 28698, line 25: Suggestion to add “-in” after zoomed, e.g. “zoomed-in”

P 28699, lines 2-5: Please provide references

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

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?

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.

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

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.

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

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.)

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?

P 28701, line 18: Suggestion to delete “stable meteorological conditions, i.e.” (unnecessary text)

P 28701, lines 20-23: I found this sentence describing line densities to be a bit confusing. 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.

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

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

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 downwind 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 observed 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.

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

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.)

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.

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

SO2 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?

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.

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.

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

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

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?

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

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

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

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

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

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

P 28707, line 5-6: This sentence was hard for me to visualize. I think it will help readability 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 horizontal wind speeds, both of which will contribute to higher estimated emission rates.)

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

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

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

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.

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?).

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?)

P 28710, lines 19-25: It may be worth mentioning previous ground and satellite based comparisons of volcanic SO2 and how your findings compare to these previous studies

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)?

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

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 K ÌĎlauea summit and East Rift emission plumes leads to a different sensitivity of the satellite instrument towards SO2 emitted at each of the two locations.

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.

Interactive comment on Atmos. Chem. Phys. Discuss., 13, 28695, 2013.