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 #1

Green: Reviewer comment.
Black: Author's reply.
Red: Modified text in manuscript.

We thank Reviewer #1 for his constructive comments and suggestions. Below we reply to them one-by-one.

Reviewer comment: 1) I think the paper is weak in the references. This is especially true for the introduction section which does not give a good overview of past studies on the subject. Instead, the authors put forward the "new potential" of satellite sensors for monitoring volcanic activity. Estimation of SO2 fluxes and lifetime from space have been reported in several papers, recently but also in early studies using TOMS data. Moreover, when referring to satellite measurements, the authors should also mention SCIAMACHY, OMI, GOME-2, IASI, AIRS (among others) in addition to the GOME-1 sensor. In the first two paragraphs of the introduction (on the role of SO2 on the atmosphere and its lifetime), it would be good to have references to key papers.

Authors' reply: We agree with the reviewer that the introduction was rather short and misses some important references and an appreciation of satellite instruments beyond GOME. We have thus extended the paper, in particular the introduction and discussion, and added some key references. However, we still aim for a brief introduction rather than a complete review of the field, as we focus on the particular conditions for Kīlauea in this case study. We thus refer the interested reader to some review papers for details and further references. We also skipped some aspects which have no relevance for our study (air quality/health).

Reviewer comment: 2) The paper is well written and the discussion section is valuable but the key message of this study is vague. Is it a paper on a technique or is it about the discrepancy between the ground- and space-based estimates of the emission rates? In the first case, the technique is not new (it is already described by the same first author in another paper) and is similar to other methods in the literature. If the focus is on the ground-based/satellite discrepancy, then the reason for it remains unexplained and the authors do not propose hints for further investigations.

Authors' reply: The paper presents emission rates and lifetimes estimated for Kīlauea. It thereby focuses both on the adaptation of the method, which is similar, but not identical, to Beirle et al., 2011, and the comparison to ground-based measurements and the discussion of the observed differences. In the revised version of the manuscript, we have strengthened both aspects:
1. We have added a discussion on the particular advantages of our approach, in comparison to other approaches in literature, to section 4.
2. We have extended the discussion of uncertainties and possible biases of both the results from ground-based and satellite measurements, and substantiate the discussion of the observed discrepancies (see also below).

Reviewer comment: It is not clear if it could be that the ground-based instruments sample only part of the SO2 plumes emitted.

Authors' reply: The ground-based DOAS measurements reported by Elias and Sutton (2012) clearly traverse the entire plume. This is evidenced by SO_2 columns reaching values compatible with null on either side of the plume traverse, as shown in their Figure 6 in Elias and Sutton (2012). Because the FLYSPEC measurements are made at a distance of only 0.3 to 1.3 km from the vent (depending on the wind direction and where the plume is intersected on Crater Rim Drive), the lateral extent is seldom more than 1 km at this point and measuring

complete transects does not pose a problem. However, due to the close proximity to the vent, the encountered SO_2 column densities and aerosol optical depths are orders of magnitude higher than those e.g. measured by satellite tens to hundreds of kilometers farther downwind, and perhaps the more interesting question is whether or not the core of the plume is adequately sampled by the ground-based traverses. Due to the high optical thickness of these plumes at close proximity, light is often prevented from penetrating the plume core. Instead, light paths that only penetrate the plume edge before being scattered towards the instrument become more likely, therefore skewing measurements towards lower column densities (Kern et al., 2012). These non-linear radiative transfer effects are now discussed in more detail in the text (Section 4.4).

Reviewer comment: Section 4 gives a description of the uncertainties of the emission rate and lifetime estimates but the latter uncertainties only relate to the method used and not to the SO2 column retrievals (see next point).

Authors' reply: We agree that the discussion of uncertainties misses the important aspects of SCDs and VCDs. We have thus extended the discussion of uncertainties considerably. In new subsections (4.2.1 and 4.2.2), we now deal with the uncertainties of SO_2 SCDs and VCDs.

Reviewer comment: 3)In view of the above, the description of the SO2 retrieval (section 2.1) is insufficient. For an SO2 plume at a presumable altitude of 1.5-2.5 km, many parameters can influence the retrieval which are not even discussed here. In reality, the results of the SO2 algorithm have not been validated and it is hard to know what it is the accuracy of the retrieved slant columns.

Authors' reply: We have expanded the description of the SO_2 retrieval, now providing the relevant information for the DOAS set-up and the calculation of AMFs, and expanded the discussion of uncertainties accordingly (see above).
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 exemplarily. 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:

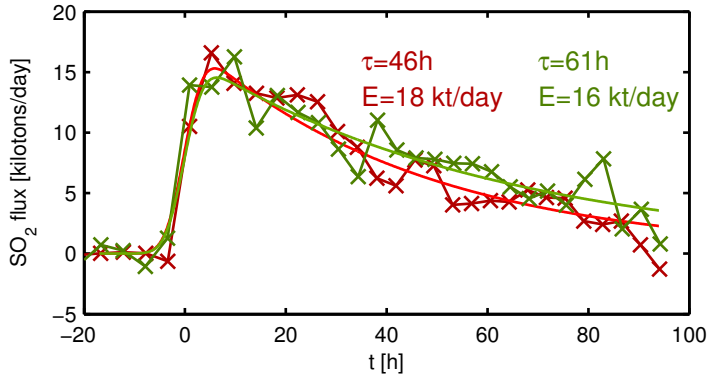

Fig. R1: SO_2 flux as function of time (crosses) and the fitted model function (smooth line) for GOME-2 (red) and OMI (green) in August 2008.

The derived lifetime based on OMI is longer by 33%, as in the OMI monthly mean the downwind VCDs 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: The exact settings for the AMF calculation are not provided either. In the 312-324 nm range used, the

wavelength dependence of the AMF is strong especially for a low plume (as it is the case here). The surface albedo used is not given, although it is arguably a large source of uncertainty on the SO2 column retrieval.

Authors' reply: We agree that our description of the method was too scarce. The extended method section now includes the respective details on the chosen wavelength (315 nm) and albedo (5%), and the discussion section now also investigates the impact of a-priori wavelength and surface albedo on the resulting AMFs.

Reviewer comment: GOME-2 is also known to suffer from several limitations at the edge of band 2 and this is not developed in the text.

Authors' reply: Due to some restrictions concerning the correction for Etalon structures of GOME-2 spectra, the lower edge of the GOME-2 band 2A (<316.5 nm) is officially not supported as "valid spectral region" (pers. communication with Rüdiger Lang, EUMETSAT, Darmstadt). The Etalon correction has been expanded to lower wavelength only since June 13, 2013, but has so far not been confirmed to significantly improve this spectral region for Level 2 data products.
In order to investigate the possible influence of Etalon structures on our SO_2 retrieval, we compared the fit residues and SO_2 SCDs over a remote area over the Pacific (145°-180°W, +-15°N) for the time period 2 weeks before and after the Etalon correction has been expanded to the lower UV. Although the overall fit quality was improved (as indicated by a decrease of the fit residues), the influence on the mean SO_2 SCDs as well as the corresponding standard deviation was found to be negligible. Additionally, data for moderately enhanced SO_2 SCDs during increased activity of Nyiamuragira volcano in June/July 2013 had been analysed with and without expansion of the Etalon correction to wavelengths <316.5 nm (by courtesy of Rüdiger Lang). Like for the Pacific area, an overall improvement of the fit could be observed. However, the corresponding SO_2 SCDs were found to be within $\approx 1\%$ of the uncorrected results.
As this is a quite complex technical detail with almost no impact on our results, we do not include this discussion in the manuscript.

Reviewer comment: GOME-2 has undergone severe degradation since its launch and it is not clear whether the elevated background values of a few kT/day observed from 2009 onwards (Figure 7) are real or not.

Authors' reply: As mentioned in Section 2.1, the detection limit for SO_2 has doubled from 2007 to 2011, and fit residues gradually increase over time. However, due to averaging (monthly means) and spatial integration (line densities), statistical errors are largely reduced (compare the vertical error bars of Fig. 4). Possible *systematic* effects of degradation on the SO_2 retrieval, on the other hand, are eliminated by our background correction east of Hawai'i. We thus conclude that the observed peak of SO_2 in 2008 and the enhanced values in 2009-2012 (compared to 2007) are real (in terms of SCDs). A qualitative comparison of our GOME-2 results to operational SO_2 data products for the OMI and SCIAMACHY instruments (available e.g. at http://so2.gsfc.nasa.gov/ and http://sacs.aeronomie.be/), which do not show that strong degradation effects, also reveal clearly increased SO_2 over Hawaii for 2008 and onwards, compared to 2007.

Reviewer comment: It is anyway in clear contradiction with the ground-based data. The authors shall expand the data description and include an error analysis to confirm (or not) if their space-based emission rates estimates are larger than the ground-based values.

Authors' reply: We have substantially expanded the retrieval description and discussion of uncertainties for both our satellite retrieval and the ground-based measurements, and substantiated our arguments why we think the satellite measurements are more accurate for this specific case, at least in 2008.

Reviewer comment: - "..spatial fluctuations, and total emissions are still highly uncertain": please provide a reference.

Authors' reply: We added a references to Andres and Kasgnoc, 1998, and to Carslaw et al., 2013.

Reviewer comment: - Please add information on the Kilauea volcano: lat,lon,elev.

Authors' reply: We added the respective information (19.4^o, -155.3^o, 1247m) to the introduction.

Reviewer comment: - All clear-sky pixels are used in the gridding procedure including the ones which contains no volcanic SO2. What is the impact of using a data filter (e.g. a cutoff value on the columns) on gridded data and hence on the lifetime and emissions rates estimates.

Authors' reply: The basic approach of this study is the estimation of SO_2 lifetime and emissions from the downwind flux. Instead of following individual air parcels, which is impossible due to the sparse coverage by the GOME-2 satellite observations, the mean flux is derived based on monthly averages. The application of a cutoff value on the noisy columns is not appropriate for this approach, as it would bias the monthly mean value for low SO_2 columns, and thereby the downwind pattern which determines the fitted lifetime.

Reviewer comment: - "..cloud altitudes derived from satellite observations have high uncertainties for low cloud fractions" : please provide a reference.

Authors' reply: For low cloud fraction, the information content on cloud heights is limited as a smaller radiance fraction actually comes from the cloud. Consequently, the impact of a-priori becomes larger, and a bias of e.g. albedo causes a bias in cloud height, which increases with decreasing cloud fraction. See Koelemeijer et al., 2003 (Fig. 4 therein).

Reviewer comment: - The plume direction is determined by the slope of a weighted linear fit. What are the weights? In this procedure, all grid pixels are accounted for, including the ones over the island with high values (Fig 2). How can it affect the accuracy of the plume direction estimate. Only one ECMWF profile is used and the threshold of 3x10e16 cm-2 make pixels far away from the volcano included in the plume direction estimation. How can it affect the results?

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: - Using horizontal transport features to derive plume altitudes have already been used in other studies. Please refer to Bluth and Carn, IJRS, 2008 and Hugues et al., JGR, 2012.

Authors' reply: We thank the reviewer for this notice, and we have added the respective references to the manuscript.

Reviewer comment: - The seasonal variation of the SO2 lifetime (Fig 5) is in contradiction with the findings of Lee et al. which show higher values in winter than in summer. The authors speculate on the role of clouds (blue crosses in Fig. 5) but they have no means really to confirm this hypothesis. The authors should test whether systematic features in their retrievals might be the cause for the observed seasonal variation.

Authors' reply: The fitted lifetimes are >40h in summer, but only 20h in spring and 30h in autumn. We observe an anti-correlation to mean cloud fraction, which shows similar "jumps" as the lifetime for April vs. May and September vs. October. We are aware that an (anti-) correlation is no proof for a causal connection. But as clouds are known to affect the SO_2 lifetime due to heterogeneous reactions, we think it is appropriate

to mention this as a possible explanation.
The seasonality is different from that found by Lee et al. over the continental U.S., but this does not have to be a "contradiction", as the SO_2 profiles (volcanic degassing at elevated vent) as well as meteorological conditions and their seasonal characteristics are significantly different for Hawai'i (tropical, maritime).

Reviewer comment: - An error on the plume height of 0.5km leads to 30% errors on the emissions. Explain why 0.5 km uncertainty is believed to be a reasonable estimate of the error on the height (and not a convenient value as it looks now).

Authors' reply: In the revised manuscript, we have extended the discussion of plume altitude. We based our estimate of the mean plume altitude on 3 different and independent sources: 1. the plume height measurements close to the vent, 2. CALIOP measurements analysed by Eguchi et al., and 3. the comparison of wind directions at different altitudes to the actual plume direction. A mean plume height below 1.5 km is unlikely, as it would be even lower than the altitude close to the vent, and the plume has high buoyancy. Also CALIOP measurements indicate rather a rising than a sinking plume. A mean plume height above 2.5 km, on the other hand, can be excluded due to the discrepancy between wind and plume directions.

Reviewer comment: - One option that has not been considered is a possible uplift of the SO2 plume after its emission. This could explain at least partly the differences between the satellite and the ground-based data.

Authors' reply: A possible uplift of the SO_2 plume during advection would indeed increase visibility (i.e. the AMF), and would gradually bias high the derived line densities. This would lead to a virtual longer lifetime, as the SO_2 decay is suppressed. The effect on E, however, would be rather low, as the maximum flux close to the source would not be affected.

Reviewer comment: - The point b in Section 4.1 simply points to choosing an appropriate lat-lon box for this study and does not bring a lot.

Authors' reply: Choosing the appropriate lat-lon box is a rather technical issue, but nevertheless necessary (otherwise the method would not work), and the sensitivity of the results on the actual choice helps to judge on what is "appropriate". We therefore decide to keep this section in the manuscript.

Reviewer comment: - "... to those reported previously at the ESA ATMOS conference (Beirle et al., 2012).." -> "... to those reported previously (Beirle et al., 2012).."

Authors' reply: The sentence was shortened accordingly.

Reviewer comment: - Page 8: (in a Lagrangian framework) please clarify.

Authors' reply: We added the following text to the manuscript:
in a Lagrangian framework (i.e., the temporal evolution of each air parcel, containing the initial SO_2 amount ΔE emitted in Δt, is considered independently, and dilution is not accounted for)...

Reviewer comment: - Please refer also to OMI. Given the better spatial resolution and noise level of OMI compared to GOME-2, it would have been a more logical choice for studying the Kilauea degassing emissions.

Authors' reply: The reviewer is right in pointing out that OMI would be well suited for a similar study. In the revised manuscript, we now refer also to SCIAMACHY and OMI. Currently, an SO_2 retrieval for OMI is implemented at MPI-C. A first comparison with the operational OMI SO_2 product results in essentially the same emission rates for August 2008 (see Fig. R1).