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Interactive comment on “A comparison of DOAS observations by the CARIBIC aircraft and the GOME-2 satellite of the 2008 Kasatochi volcanic SO₂ plume” by K.-P. Heue et al.

K.-P. Heue et al.

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Received and published: 15 April 2010

Authors Comment:

We thank the referees for the extensive review and all the helpful comments.

Interactive comment on "A comparison of DOAS observations by the CARIBIC aircraft and the GOME-2 satellite of the 2008 Kasatochi volcanic SO₂ plume" by K.-P. Heue et al.

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Received and published: 8 March 2010

Paper describes near simultaneous sampling of the drifting volcanic SO₂/aerosol volcanic cloud by CARIBIC in-situ aircraft laboratory and satellite GOME-2 UV spectrometer. The aged cloud originated from August 8 2008 Kasatochi and the sampling occurred week later on August 15 over Europe at altitude 11 km. Aircraft encounters with volcanic clouds are quite rare; the previous one was of NASA DC-8 research aircraft inadvertently flown into aged volcanic cloud from Hekla eruption in 1999 [Rose et al 2003]. The DC-8 measurements were compared with satellite UV Total Ozone Spectrometer SO₂ and Aerosol Index measurements. The limiting factor in TOMS comparisons were high solar zenith angles and limited TOMS sensitivity to volcanic SO₂.

[Rose, W.I., Gu, Y., Watson, I.M., Yu, T., Bluth, G.J.S., Prata, A.J., Krueger, A.J., Krotkov, N., Carn, S., Fromm, M.D., Hunton, D.E., Viggiano, A.A., Miller, T.M., Ballentin, J.O., Ernst, G.G.J., Reeves, J.M., Wilson, C. and Anderson, B.E. 2003. The February-March 2000 eruption of Hekla, Iceland from a satellite perspective. In: Volcanism and the Earth's Atmosphere (eds. A. Robock and C. Oppenheimer), AGU Geophysical Monograph 139, pp. 107-132, 2003]

The Kasatochi eruption occurred at fortunate time when multiple research and operational satellite platforms were in operation (MetOp launched in 2006, EOS Aura launched in 2004 and EOS Aqua launched in 2002) carrying IR and UV satellite instruments capable of detecting volcanic gases and aerosols with unprecedented precision not possible just few years ago. On the other hand, the DOAS SO₂ measurements from aircraft platform are new and to my knowledge this is the first such measurement of the volcanic cloud. Authors specifically compare satellite DOAS SO₂ data from GOME-2 UV instrument with aircraft DOAS SO₂ measurements from CARIBIC platform, few hours after GOME-2 overpass. TRAJKS trajectory model was used to account of cloud

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advection and correct for the differences in observational times. The observing conditions were quite favorable with solar zenith angle $\approx 73^\circ$. Good agreement between GOME-2 and CARIBIC DOAS SO₂ data is encouraging as it indirectly validates both retrievals. The only addition on my "wish list" would be adding in-situ SO₂ instrument to CARIBIC payload.

I recommend publishing the paper in ACP with minor corrections aimed at improving the text.

General comments:

1) I suggest adding in-situ SO₂ detector to CARIBOC payload, which would validate DOAS SO₂ measurements (e.g. Luke, W.T., 1997. Evaluation of a commercial pulsed fluorescence detector for the measurement of low-level SO₂ concentrations during the gas-phase sulfur intercomparison experiment. Journal of Geophysical Research 102(D13), 16255-16265.)

The referee is definitely right that the CARIBIC project would benefit from a high sensitive in situ SO₂ instrument. However there are not many SO₂ in-situ instruments suitable for airborne measurements available (e.g. Speidel et al., 2007).

Additional ideas for airborne in-situ SO₂ measurements are highly welcome. We currently think of building an adequate instrument for the integration on CARIBIC.

An according statement is included in the conclusion.

2) The text is not always clear. English needs to be improved throughout. Some suggestions are given in detailed comments.

Thank You for suggesting the improvements.

3) Radiative transfer model and calculation of box-AMFs (e.g. shown in figure 1) need to be described.

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An according section is included in the manuscript in section 2.2, description of the CARIBIC DOAS instrument, and the data retrieval.

4) Specify the error in CARIBIC caused in SO₂ retrieval by constant temperature 273K (p.528) assumption. This temperature is not realistic for likely SO₂ plume altitude and is also inconsistent with ozone cross section temperatures in the DOAS fit (223K and 243K).

According to the temperature measurements of aeroplane the best temperature would be 223K. The systematic error introduced by the incorrect temperature of the SO₂ cross section is about 6% in the wavelength range 312-330nm. For the comparison however, this error is less important as the same error is made for both instruments. An according section is added to the description of the cross sections.

5) Do adjacent cross-track GOME-2 pixels overlap? If so, show actual GOME-2 pixel shapes in figure 8,9 and 11.

GOME-2 Pixels do overlap, therefore the figures are changed.

6) I found it surprising that GOME-2 AMF only increases by 10% when thick cloud (COT 10) is placed just below SO₂ layer (p539, line 11-12). Suggest increasing cloud single scattering albedo (SSA) from 0.99 to 0.9999 and re-calculating AMF.

We were astonished ourselves, when we did the calculation, but if you add a highly reflecting surface at this altitude (11km), which is more or less what you do when including the clouds, why should that increase the light path within the next km compared to the geometrical approximation? The air density at 11km (205hPa) is so low that the high reflectivity and hence increased intensity of the cloud hardly affects the weighting function at this altitude. A bigger difference relative to the cloud free

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case can be observed at lower altitudes ≈ 5 km or at ground level if the assumed albedo is changed.

We recalculated the AMF for the dense cloud (COT 10) with the enhanced SSA as the referees suggested, with no aerosols above the cloud. For the lower SSA (0.99) used before the AMF increased between 5-7% compared to the geometrical approximation, only a slightly stronger increase (upto 12%) was observed when a highly reflecting cloud (SSA=0.9999) is used instead.

The discussion of the AMF is broadened by some of these aspects.

7) I found surprising assumption of highly absorbing aerosols with SSA=0.8 in volcanic cloud. The predominant aerosol component in week old volcanic cloud should be sulphuric acid droplets with SSA=1. Provide more evidence supporting this assumption.

According to Martinsson et al. (2009) the aerosols contain a large fraction of carbon:

The aerosol sample collected seven days after the eruption contained a substantial carbonaceous fraction, being a factor of 2.3 higher in mass concentration than particulate sulfur (CS). It also contained a component probably originating from volcanic ash represented by silicon (0.07CS) and iron (0.03CS), which was significantly depleted already a month after the eruption. The relative importance of the carbonaceous component declined to on average 1.0CS 3 - 4 months after the eruption.

Usually high carbon content in the aerosols causes a dark aerosol (low SSA). Moreover the observed intensity decreased during the period of the plume observation (Figure 4), according to our radiative transfer calculation this decrease can not be explained with highly reflective aerosols (SSA $\gg 0.99$). Both these observations indicate that the SSA is less than unity.

As the assumption on the SSA is already based on the findings by Martinsson et al. (2009), no changes are made in the manuscript.

8) To achieve good agreement between trajectory shifted CARIBIC SO₂ columns and GOME 2 SO₂ spatial distribution, the wind speed in trajectory model has to be locally enhanced by 25%. To test this hypothesis I suggest forward project trajectory for the next 3 hours and compare with OMI overpass at 12UTC (see OMI figure). Perhaps, comparing GOME-2 and OMI spatial SO₂ patterns would help constrain local wind speeds.

Comparing the plumes position also with OMI data is a logical consequence of the performed comparison and the findings about the trajectories. Therefore we thank the referee for this comment. The AURA satellite passed over Europe at 11:58 UTC, hence 6 hours after the CARIBIC observations and 3 hours after GOME-2.

However the comparison with the OMI data (http://disc.sci.gsfc.nasa.gov/Aura/data-holdings/OMI/omSO2_v003.shtml) showed that, neither the ECMWF based Trajks model nor the simple model based on the measured wind speed and the local wind direction, lead to a satisfying agreement with OMI's spatial pattern. The wind speed optimized for the GOME-2 observation was too high and in the TRAJKS model the wind speed was too low. But also the position of the main plume does not match that good as it did with the GOME-2 data, so it seems that the wind field in that region is far more complex as assumed in ECMWF data and the assumption of constant wind speed and wind direction is not valid for a time period of 6 hours.

A respective section and a figure are added to the manuscript in Sec. 4.3.

Specific comments:

524, 11 "A comparison of the [satellite] spatial pattern with . . ."

Done

14 "[Emitted] and secondary particles,. . . " - Emitted volcanic ash particles should have fallen out after a week of travel.

*We do not agree with the referee in this point, according to Williams et al. 2002 the lifetime of particles larger than 50 nm exceeds 7 days. Martinsson et al. 2009 found clear evidence for volcanic ash in this aerosol plume (see answer to general remark 7)
No changes were made to the manuscript*

17 suggest re-wording: The main remaining sources of error are uncertainties in local wind speed . . . and effects of aerosols on DOAS retrievals.

Done

20. I suggest adding SO₂ in-situ instrument to CARIBIC payload

good idea - see general remark 1

525 11 "several satellite [UV spectrometers] , e.g." - explain abbreviations, provide references

Done

17 remove [here]

Done

20 Suggest re-wording: "Because of slight inconsistency between GOME-2 and CARIBIC SO₂ retrievals a more detailed study of local wind pattern was performed, which resulted in better agreement"

Done

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24. Why is O_4 slant column mentioned here? What is O_4 relation to SO_2 column ?

The oxygen dimer O_4 is typically used as indicator for clouds and aerosols in the specific wavelength range (320-380 nm). The clouds also affect the AMF / WF and thereby also the SO_2 slant column.

Some more details on this topic are included in the radiative transfer section.

526 6 Why in-situ SO_2 is not measured?

Currently no in-situ SO_2 instrument is installed on CARIBIC, but it is planned to install one. See comment 1).

7 [in] real time

Done

25. "pointing starboard" - not clear. Are telescopes pointing toward or perpendicular to the flight direction? Will be nice to have photo of the telescopes on the aircraft.

More details are given here, since the telescopes are very small the viewing direction can hardly be seen on any photo of the pylon, additional details can be found in Dix et al. (2009)

527, 4 "full width [at] half maximum"

Done

10 "which" -> "that are "

Done

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10 "weighted average light path" - weighted with what?

"Intensity weighted" - corrected

13 Suggest re-wording: "The sensitivity [of the measured spectral radiance] to a trace gas concentration at certain altitude is commonly known as box air mass factor (box-AMF) or weighting function (WF)".

Done

25. What was the SZA during plume encounter?

During The CARIBIC observation the Solar Zenith Angle varied between 76.4° and 73.2° as it was mentioned in the context of the AMF simulation. For the GOME-2 observation (4 hours later) the SZA was 44° , this information was not mentioned before and is added for the calculation of the geometrical AMF (page 531 line10).

26 "For comparisons with other observations [and models] the vertical column density is used"

Done

29 "is called air mass factor [or column weighting function] "

Done

Throughout the manuscript the expression AMF is used, here for clarity the column weighting function is included.

528, 1 Box AMF also depends on clouds and aerosols. Suggest re-wording: ". . . but also on the gas vertical profile shape"

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Done

6 " O_4 observations are often used as proxy to estimate cloud properties" - This statement is not clear: what O_4 observations (in-situ density or absorption, etc) are meant and what cloud properties (e.g. optical or physical) can be estimated? Also why O_4 , but not for example O_2 observations are used?

Additional details are given in this section. The referee's suggestion to use O_2 absorption in addition to O_4 can not be accepted, since the next O_2 absorption bands are either below 260nm or around 628 nm and hence not in the interval observed by the CARIBIC DOAS instrument. Even if the CARIBIC DOAS wavelength interval was including the 628 nm line, the influence of aerosol and cloud would be different in this spectral range.

more details are given.

12. "ultra violet" - one word, often UV

Done

20 For SO_2 the same cross section (273 K) as for the GOME-2 data retrieval is used" - This is certainly not correct temperature for the CARIBIC cruise altitude, so absolute SO_2 column density will be incorrect. Specify the systematic SO_2 error.

The systematic error of the incorrect temperature is about 6%. Included, see general comment 4)

531, 2 Suggest additional reference here "In such cases this non-linearity has to be corrected for [Yang et al 2009] , or . . ." Yang, K., N. A. Krotkov, A. J. Krueger, S. A. Carn, P. K. Bhartia, and P. F. Levelt (2009) Improving retrieval of volcanic sulfur diox-

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ide from backscattered UV satellite observations, *Geophys. Res. Lett.*, 36, L03102, doi:10.1029/2008GL036036.

Included next to the reference Richter et al., 2009.

18. Add web link for NRT SO₂ data from both OMI and GOME-2 .
[http://satepsanone.nesdis.noaa.gov/pub/OMI/OMISO₂/](http://satepsanone.nesdis.noaa.gov/pub/OMI/OMISO2/)

Done

21 suggest: ". . . are shown"

Done

23: suggest re-wording: ". . . an eastern edge of the volcanic cloud has reached Western Europe stretching from Western Mediterranean to the Baltic Sea."

Done

532, 23 Explain how COT was determined. 25 How single scattering albedo value 0.99 was chosen?

The COT was determined by comparing the O₄ SCD with the results of the AMF calculation, also the intensity relative to the cloud free reference was considered. The single scattering albedo was chosen because with higher values the decrease in the intensity observed during the first cloud would not be possible at all. Besides the aerosol number concentration increased when the plane descended to the cloud, and according to our previous comments on the SSA of the aerosols above the clouds these aerosols are rather dark, thereby the SSA of the cloud droplet aerosol ensemble is lower than for typical clouds.

The general description is included in sec. 2.2

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533,7 *"The sensitivity to local SO₂ concentrations [at flight altitude] is enhanced"*

Done

15. *suggest re-wording "the difference in [SO₂] SCD peak heights can be explained by different SO₂ columns"*

Done

22. *cloud cover " -> cloud layer*

Done

29 *"gives reason to assume" -> suggests*

Done

534, 4 *re-word: "we assume the plume altitude between 11 and 12 km"*

Done

20 *"by which" -> therefore*

Done

537, 7 *re-word "A good agreement between the trajectory projected SO₂ timeseries and GOME-2 measurements is found with an 25% increased wind speeds "*

Done

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11-12 remove "the spatial patterns of"

Done

538,16 " . . . with single scattering albedo [SSA=0.8]. . .' - How the SSA was estimated?

The single scattering albedo of the aerosols as well as the aerosol extinction were estimated from comparison with the observed intensities at 320 and 360 nm. (See Comment 7)

539, 11-12 "the sensitivity for GOME-2 increases from 2.19 to 2.4 for nadir when the dense cloud is assumed, instead of the optically thin one" - The satellite AMF should increase more for an SO₂ layer just above dense cloud with COT=10. What cloud fraction was assumed in AMF calculation?

Based on the pictures of the on board video camera we assumed a completely dense cloud layer i.e. 100% cloud cover.

See general comment on the increase of AMF.

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