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Atmospheric Measurement Techniques Discussions

Interactive comment on "Early in-flight detection of SO₂ via Differential Optical Absorption Spectroscopy: a feasible aviation safety measure to prevent potential encounters with volcanic plumes" *by* L. Vogel et al.

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First of all, the authors would like to thank the reviewers for their constructive comments and suggestions improving the manuscript. In the following, first the general comments of the individual reviewers together with their minor comments are addressed individually. Changes to the manuscript based on both reviewers' comments are given at the end of this answer.

1 Reviewer R. Campion

I've read with interest the paper entitled "Early in-flight detection of SO_2 via Differential Optical Absorption Spectroscopy: a feasible aviation safety measure to prevent potential encounters with volcanic plumes." The paper is clearly written and well structured and is certainly an important and innovative contribution to the boiling hot problem of volcanic plume hazard for the aviation safety. It examines the ability to detect SO_2 by a cluster of onboard, forward pointing mini-DOASes and to use their information to avoidthe plume by changing the flight altitude. SO_2 is used here as a proxy for volcanic ash, usually the most dangerous compound of the plume. Based on plane approaches of the SO_2 rich plume of Popocatèpetl volcano (Mexico), as well as sophisticated radiative transfer simulations, the authors convincingly demonstrate that this technique works very well and at great distance for good viewing visibility. Furthermore, the wavelength dependent attenuation coefficients of SCDs reported in this study will be of interest for the DOAS users among the volcanology community. For these two reasons this paper is a valuable contribution that deserves to be published in AMT. I would like however suggesting a few additions and corrections that could improve the paper before its publication.

My main concern is about the applicability of the method in the case of an ash rich plume. The ash plume of Popocatépetl was ash free, while the plumes that are the most dangerous to airplanes are ash rich. I understand that a field campaign with ash and SO_2 plume encounters is difficult and costly to set up, but I think that some radiative transfer simulations taking into account ash in the plume should be added to the paper. This might be done for example by adding a collocated scattering and absorbing aerosol in the model run type B, supposed to represent a large scale volcanic plume. This point is important to because Kern et al. (2009) have documented the attenuation effect of ash on SCDs measured using UV cameras measurements. The

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presence of ash could partly mask the SO_2 absorption and diminish its detectability by UV sensors.

Authors' comment: To address the concerns raised, an additional radiative transfer study has been performed denoted as model run (B2), which assumes an ash laden cloud consisting of aerosols with a single scattering albedo (SSA) of 0.8. This value is at the lower end of reported ones (Prata and Grant , 2001; Pavolonis et al., 2006; Kudo et al. , 2008) resulting in a possible underestimation of the simulated SCDs and thus acts as a lower limit. The results have been plotted in an additional figure (Fig. 1). Additions were made to the manuscript to describe this model run in the experimental section and in the results, also given in the section of general changes to the manuscript in this answer to the reviewers.

The second point that could be improved in the paper is the comparison with the existing multispectral IR imaging system introduced by Prata and Bernardo, 2009. Does it have a longer detection range? Are there cases when UV detection works where IR could fail? Are the two methods complementary (remote detection vs. close range 2D imaging?)

Authors' comment: Both methods are complementary and detection ranges of both methods are comparable assuming best conditions for both systems. For 2D IR imaging systems, a detection range of 100km has been reported based on theoretical considerations (Barton and Prata , 1994). The manuscript has been modified to better reflect that these systems are complimentary (see list of changes at the end). The main advantage of the UV system lies in lesser interference with water. Even if negligible ambient water vapour is present in the atmosphere at higher cruising altitudes, one might expect water vapour of volcanic origin or carried from lower altitudes together with the volcanic cloud. Once an improved DOAS system has been developed, which is tuned to early detection of SO_2, a campaign involving portable IR systems as described in Prata and Bernardo (2009) would be the next step.

Minor Comments:

In page 2831 I think the authors could mention the paper by Bernard and Rose (1990) about the crazing effect of H2SO4 on the airplanes windows, which is more detailed that the ICAO report cited in the manuscript.
Authors' comment: Indeed the reference describes the effects in much greater detail. It has been added as an additional reference.

Page 2833 ln 10: replace identify by identified
Authors' comment: done

Page 2841. Although I'm not a native English speaker, the last sentence of paragraph 3.3 sounds odd to me and should be reworded.
Authors' comment: The sentence has been replaced by: *In this way, the error reflects uncertainties of measurements at greater distance to the plume.*

Page 2846: replace e.q. by e.g.
Authors' comment: done

Page 2855 The use of the x symbol for "times" might create some confusion with ln(X) and its Taylor approximation. Also the term "weak absorber" needs to be defined explicitly.
Authors' comment: To prevent confusion, x has been replaced by ξ in the equation. In the subsequent sentence, *weak absorber* has been clarified by *weak absorber with optical densities on the order of a few percent.*

Caption of figure 6: replace an vertical by a vertical

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Authors' comment: done

2 Reviewer Anonymous Referee

In their manuscript "Early in-flight detection of SO_2 via Differential Optical Absorption Spectroscopy: a feasible aviation safety measure to prevent potential encounters with volcanic plumes", L. Vogel et al. report on airborne measurements of SO_2 from a volcanic plume using the DOAS method. Several approaches of the plume are documented and compared to stationary and mobile ground-based DOAS measurements. The airborne measurements are simulated by a radiative transport model and extrapolated to observations from larger distances to the plume. Some additional RTM calculations are made to investigate the impact of larger optical depths (from SO_2 and aerosols) on the observations. From their measurements and the simulations, the authors conclude that passive UV DOAS measurements could be used on aircrafts to avoid flying into volcanic plumes.

The paper is well written and reports on interesting measurements. The topic is within the scope of AMT and I find the test case and the idea of applying UV DOAS instruments to operational volcanic plume avoidance intriguing. However, while the test measurements and their comparison with ground-based data are sound if somewhat qualitative, the discussion of the application to volcanic plume avoidance is not convincing. In my opinion, additional RTM studies and discussion are needed to justify the title of this manuscript, and therefore I can only recommend publication after major revisions.

My main concern about this manuscript is that it claims to have shown that passive UV DOAS measurements from aircraft can be used for volcanic plume avoidance while in fact it has only demonstrated that SO_2 plumes from volcanic plumes can be detected at relatively large distance when flying exactly in the altitude of the plume. While this is

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a nice demonstration, it is not really surprising as SO_2 has been observed before from airborne DOAS instruments in volcanic plumes and in power plant emissions.

Authors' comment: DOAS measurements of SO_2 have been performed for a long time and from various platforms, including aircraft. Usually studies have been performed relatively close to the volcanic plume/cloud and/or under different radiative transfer conditions (e.g. satellite measurements), but maximum detectable distance of observer to volcanic cloud has never been subject of a study. The fact that SO_2 is observable is not surprising, nor that it is detectable from a certain distance. However, the DOAS technique used for aviation safety by introducing a forward looking instrument on aircraft with real-time evaluation algorithms is a novel application, to which assessment of maximum detectable distance is of essence.

To make such a system useful for volcanic plume avoidance, a couple of requirements must be met:
1) The system must tell the pilot at which distance and in which altitude a dangerous SO_2 plume is observed. It is not clear to me, how the distance to the plume can be estimated from the DOAS measurements alone, unless some kind of triangulation is applied which does not appear very realistic to me. Also, how is the altitude of an extended plume estimated from the measurements? This is crucial information for any attempt to avoid the plume. Measurements under different angles are potentially a method to estimate the plume altitude, but again this is complicated by the fact that the distance to the plume and also its SO_2 content are not known. Please explain in the manuscript how plume height and distance can be determined from the measurements of the instrument.

Authors' comment: The information given by an early detection system should include the exact location of a volcanic cloud and distance to the aircraft in the best case. For passive systems as UV based DOAS presented in this study as well as camera systems in the IR, assessing the distance to the plume is very difficult and probably includes RTM with high computational needs unsuitable for real time evaluations. However, for successful avoidance of the volcanic cloud, it is most important to determine if the cloud is in the line of flight of the aircraft. So far, the angle at which the plume is detected is the best approximate that can be given which is satisfactory for the pilot in order to take evasive action. A fully operational system would need to include a multitude of viewing directions, improving detection and leading to improved advise to the pilot. In the scope of this paper, we can only explore the feasibility of the method rather than its fully integrated implementation.

2) The system must be sensitive enough to give a warning when there is still enough time to change course. While the paper contains some discussion on this, I think that an estimate of the smallest observable OD must include the dependence on illumination (flight altitude, solar zenith angle, solar azimuth angle) and also give some indication on how the background reference is to be taken in an automated system. Using a measurement from just after passing the plume is not an option in real world applications, and other alternatives (fixed background, zenith-sky observation from another telescope / instrument / stripe on CCD) have negative impacts on the detection limit. Considering that even with the rather optimistic assumptions made in the current manuscript, there only are a few minutes between the first measurement above detection limit and contact with the plume, this is a relevant discussion and should be included in the manuscript.

Authors' comment: The reviewer is absolutely correct that there are many issues to be solved before proposed system can be regarded as mature. Here the authors report only a "proof of concept" study. To improve the manuscript, a part has been included in the conclusion (see below) that addresses these issues and mentions possible directions of research. Estimates on smallest observable OD including dependence on flight altitude, solar zenith angle, solar azimuth angle and probable cloud cover are beyond the scope of this manuscript but rather a publication in itself.

For the construction of the background reference, one could think about e.g. default Fraunhofer reference spectra (FRS) constructed from a high resolution solar spectrum including dependencies on altitude, SZA, SAA, instrumental issues, etc. These could be loaded from a small database to fit the conditions at hand. Also, a measured spectrum could be used, e.g. taken every 30min if no absorptions are present, an additional telescope pointing at zenith to measure a spectrum to which the FRS for the forward direction are scaled/constructed. There are many possibilities, but how to exploit them depends strongly on possible future measurement systems.

3) The system must be able to differentiate between a dangerous SO_2 *plume at flight altitude and a harmless* SO_2 *plume above. As the light observed by the forward viewing telescopes is mostly scattered at flight altitude, any* SO_2 *layer above the aircraft will also create a signal (depending on SZA). To a much smaller extent this is also true for* SO_2 *at levels below flight altitude. I think that RTM calculations with plumes of similar* SO_2 *content but at different altitudes are needed to investigate their impact on the signal. In addition, there are some obvious drawbacks of using passive UV DOAS instruments for volcanic plume detection which should be briefly mentioned in the conclusions, for example the fact that* SO_2 *is measured but ash is more dangerous, that measurements can only be performed at daylight, and that clouds can interfere with the observations.*

Authors' comment: A SO_2 cloud above the aircraft will indeed lead to an increased signal of SO_2 for all measured angles. How to account for this is one of the challenges to be solved if this method should be applied on a regular basis on commercial carriers. An additional telescope looking at zenith would be one solution, others might be based on algorithms calculating a corrected SO_2 image from the measured 2D-SO_2-distributions to still guide the pilot into the least hazardous direction. RTM calculations for plumes of similar SO_2 content at different altitudes would indeed be very interesting, but again this would be beyond the scope of the current manuscript. To raise more attention to these issues of passive SO_2 detections by DOAS, they are now highlighted

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in more detail in the conclusion.

Minor comments

P2834, l11: reported appears twice
Authors' comment: Deleted the first "reported".

P2834, l26: shouldn't that be 1.730 Gg / day?
Authors' comment: Absolutely

P2835, 14: a recent examples ⇒ recent examples
Authors' comment: Done

P2835, l23; as sketch ⇒ a sketch
Authors' comment: Done

P2836, l6: Why "thus"? This is a different aspect
Authors' comment: Thus has been deleted

P2837, l14: shouldn't this be 1.9 Gg / day?
Authors' comment: Absolutely

P2845 l24: are observed are increasingly ⇒ are observed increasingly
Authors' comment: Done

P2848, l2: thus the all modelled ⇒ thus all modelled

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Authors' comment: Done

P 2849, l25: what are the units of epsilon?
Authors' comment: Units of epsilon is 1/length. The results in this study are given always in 1/m. Next to P2849, l25: ...7.33×10^{-5}m^{-1} , the unit is now stated in table 4 and 5.

P2851, l11: I think that intensity is also very important for the detection limit
Authors' comment: The authors chose to give the detection limit in optical density, because this unit does not take into account any instrumental issues. Commercially available higher grade spectrograph with low stray light are easily capable of measuring these optical densities. Low intensity due to the optical set-up or high SZA can be countered by higher exposure times up to a certain degree. Intensity issues due to measurements only possible during daylight are discussed in in greater detail in the revised manuscript.

Fig. 3 4: please use the same scale
Authors' comment: Figure 4 has been adjusted to have the same scale as fig 3.

Fig 5: I assume this is the absorption cross-section and has units of cm^2 / molec
Authors' comment: Done

Fig 7: This is probably not intensity but differential optical density
Authors' comment: Done

Fig 13: caption: concentration is molec / cm^3, not molec/cm^2

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Authors' comment: Done

Fig 14: Check first sentence in caption for grammar
Authors' comment: The sentence has been changed to *Optical densities are shown for model scenario B in order to assess detectability of a large scale volcanic cloud.*

3 Changes to the manuscript based on both reviewers' comments

In the following, a detailed account is given on how the manuscript has been changed based on the reviewers' comments and suggestions. Changes in the manuscript are given in the order they appear.

1 Introduction

P2833, L28: In order to keep the manuscript up-to-date and stress the point of possible separation of SO_2 and ash cloud, a reference to Thomas and Prata (2011) is included. Also the sentence was added *Even if most of the ash and* SO_2 *have separated, the* SO_2 *cloud might still contain fine ash particles (Thomas and Prata , 2011).*

P2834, L8: A reference was added to give the range of IR imaging systems (Barton and Prata , 1994). The sentence was appended: *The maximum detection range of such a system is 100km (Barton and Prata , 1994).*

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3.4.2 Model Runs B: Large scale SO_2 clouds

P2842: Additional radiative transfer studies have been done to clarify the effect of different amounts of ash in the volcanic clouds. They are denoted model runs B2 and a description of their set-up is appended. The text has been modified to:
... To test the sensitivity of such instruments to large scale volcanic SO_2 clouds, model runs B were set up using a SO_2 cloud with infinite extent in one horizontal direction at 10 km altitude. A SO_2 concentration of 1×10^{12} molec cm-3 was assumed for the simulation. Aerosol particles are simulated in model run B1 as in model runs A as purely scattering with a single scattering albedo of 1, a Heyney-Greenstein asymmetry parameter of 0.8, assuming scattering of sulphate aerosols. The cloud exhibited an AEC of 0.1 km-1. Additionally model run B2 is performed which simulates different ash contents of the cloud. The ash is assumed to have a single scattering albedo of 0.8, which can be regarded as a conservative estimate for single scattering albedo of ash (Prata and Grant , 2001; Pavolonis et al., 2006; Kudo et al. , 2008). The varying ash contents are studied by assuming different AEC of 0.1km-1, 0.5km-1, 1km-1 and 4km-1. The SO_2 concentration is the same as in B1. ...

5 Comparing measurements to model results . . .

P2851: Results of the additional radiative transfer run with ash aerosols in the volcanic cloud have been included in the section and a figure showing the results was added. Next to minor adjustments in the paragraph in order to distinguish between model runs B1 and B2, the following text was added:
The results of B2 show the system's response to varying ash contents of the volcanic cloud. Ash was simulated by decreasing the single scatter albedo (SSA) of the plume aerosol to 0.8. This is thought to represent a lower limit for the SSA of an ash-rich volcanic cloud (see e.g. Prata and Grant , 2001; Pavolonis et al., 2006; Kudo et al.

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, 2008). The modelled optical densities obtained for 310.8nm are shown in Fig. 15, as the highest sensitivity was obtained at this wavelength for model run B1. For comparison, the result of B1 (Fig. 14) for 310.8nm is depicted as a dashed blue line. Reducing the SSA from 1 to 0.8 at an aerosol extinction coefficient (AEC) of 0.1km^{-1} reduces the sensitivity very slightly, decreasing the detection range from about 90km to 85km. However, a further reduction in signal is observed for optically thick plumes, as the light path inside the cloud is reduced. Moderate AECs of $<$1km^{-1} lead to a decrease of the maximum distance of detection from $>$80km to about 60km. A very thick cloud with an AEC of 4km^{-1} would only be detectable from ≈35km distance. However, such a cloud would be clearly visible in the sky, as the scattering extinction length is only 250m. While such conditions may be encountered in close proximity to the volcanic source, they are not typical of a large-scale, diluted volcanic cloud that has travelled many tens or hundreds of kilometres in the atmosphere.

An additional Figure has been added to visualize the results (Fig. 1). Its full caption is: *Volcanic ash is simulated with a SSA=0.8 and varying AECs. For comparison, the dashed blue line shows the result for a cloud with a purely scattering aerosols at an AEC of 0.1 (see Fig. 14). The dashed black line represents the assumed detection limit of 10^{-3} in optical density. Higher ash content reduces the detection limit, but a detection range of > 60km is still obtained for AEC < 1. Only for extremely thick plumes, such as those encountered in close proximity to the volcanic source, does the detection range drop further.*

Conlusions

P2851-2852: Reflecting the concerns of the reviewers, changes were made to the conclusion also including the new findings from the additional model run B2. The paragraph was changed to:

The measurements presented here clearly demonstrate the general applicability of DOAS as an early detection technique for SO_2 *in a "proof of concept" campaign. A number of plume approaches were flown, and the measurement results were reproduced with a radiative transfer model. Although the approaches were only started at up to 25 km distance to the plume, the found relationship of signal to distance of the measurements could be used to extrapolate the experiment to 100 km distance. Due to the lower air pressure at typical flight altitudes (about 10 km) when compared to the altitude of the Popocatépetl plume, additional radiative transfer studies conclude that a volcanic plume with a* SO_2 *slant column density of* 10^{18} *molecules cm*$^{-2}$ *as viewed from the outside can be detected at distances up to 80 km away for both, a cloud consisting only of purely scattering sulfate aerosols (SSA=1) and a cloud consisting of ash with a SSA of 0.8 and aerosol extinction coefficients typical for a large scale diluted volcanic cloud. This range provides enough time for pilots to take actions to avoid plume fly-through under typical flight conditions, suggesting that this technique can be used as an effective aid to prevent dangerous aircraft encounters with potentially ash-laden volcanic plumes.*

However, certain issues must be addressed. Because the technique is based on radiation in the UV spectral region, it is only applicable during daylight. At twilight the signal to noise ratio will drop due to reduced intensities in the UV, which can partly be compensated by longer exposure times with consequent lower measurement frequency. The technique does not detect the main hazard volcanic ash. Although certain algorithms have been proposed which should be able to determine aerosol optical densities of the plume from UV measurements (Kern et al., 2010b), these concepts need a high computational power and are not feasible for real time evaluation. SO_2 *is only a good proxy for ash which is the greater hazard if there is no separation of the ash and* SO_2 *cloud. Thus the technique is only complementary to ash detection systems in the IR.*

One has to keep in mind that this study is only "proof of concept" and does not present a mature system. Further efforts are needed in experiment and modelling to fully explore the capabilities of the technique. This includes the ability to spatially resolve

volcanic plumes at greater distances in order to allow avoidance measures to be initiated. Strategies must be developed to supply clear sky reference spectra without SO_2 *absorptions. One approach would be to construct a small database with reference spectra constructed from high resolution solar spectra (e.g. Chance and Kurucz , 2010) including dependencies on altitude, solar zenith and azimuth angle. Furthermore, algorithms need to be developed which reduce or eliminate the influence of a* SO_2 *cloud above or below the aircraft which could influence the perceived signal at all viewing directions. Also investigations of the limitations e.g. in case of high altitude clouds between plume and instrument need to be done. This includes sensitivity to a volcanic cloud with ash particles covered in ice need to be addressed, because this is one of the cases were IR techniques based on the reverse absorption method are not suitable.*
Last but not least, great potential lies in the development of DOAS instruments developed to this specific task. Large volcanic clouds are much more easily evaded by flying over or under them than by trying to go around them. Therefore, the vertical direction is arguably more important than the horizontal one. E.g. one could imagine a DOAS instrument applying an imaging spectrometer, which could be positioned so that its spatial axis is in the vertical, its dispersive axis is horizontal (IDOAS, Louban et al., 2009).
Besides the limitations and need for future research mentioned above, DOAS based SO_2 *detection is a complementary technique to the detection of ash in the infra-red regime and in combination can greatly mitigate the risk from volcanic clouds to aviation.* Concluded

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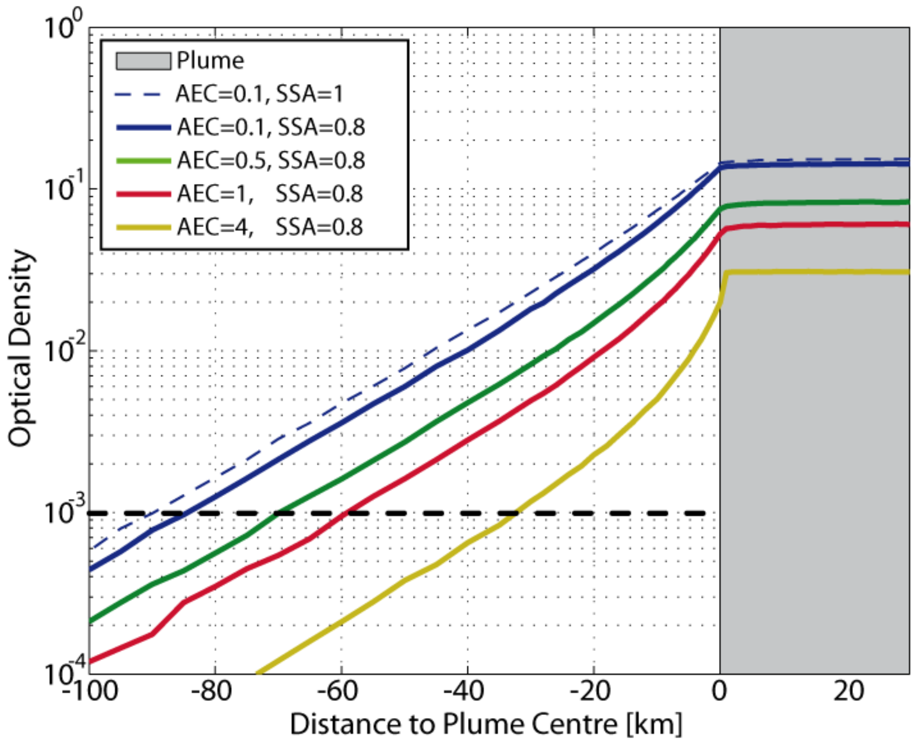

Fig. 1. Optical densities for model scenario B2: Depicted are Simulated Optical densities at 310.8nm as a function of distance to an extended volcanic cloud (scenario B2) and varying ash content.

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