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Atmospheric Measurement Techniques Discussions

# *Interactive comment on* "iFit: An intensity based retrieval for volcanic $SO_2$ from scattered sunlight UV spectra" *by* Ben Esse et al.

**Anonymous Referee #1**

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The manuscript "iFit: An intensity based retrieval for volcanic SO2 from scattered sunlight UV spectra" by B. Esse and co-authors present an alternative method for the evaluation of SO2 column densities from of UV/vis spectra. In contrast to classical DOAS, their method does not require a measured reference spectrum, which may display a crucial limitation for measurements at volcanoes when the plume contaminates the entire field of view of the instrument.

The topic is suited to be published in AMT. The manuscript, however, requires major revisions as detailed below. The overall goal should be that the method is formulated as clearly as possible to allow fellow scientists to reproduce the results.

* Major comments *



1) In the Abstract (l. 5) the authors claim to present a novel technique using intensity fitting and a solar reference. This claim, in my opinion, is not fulfilled because, in general, intensity fitting is equivalent to fitting in optical density space and commonly applied in the IR community. Secondly, using a literature solar reference was already applied by Salerno et al., 2009b and discussed more thoroughly by Lübke et al., 2016. What makes the described approach interesting is the calculation in high resolution. This detail, however, is not motivated enough in the abstract/manuscript. Why should it be advantageous to fit in high resolution while the actual measurement spectrum does not provide any information in this increased resolution anyway?

I would like to take this opportunity to mention that there are numerous publications addressing the correction of simplifications introduced by DOAS. These were compiled by Referee #1 in his/her comment on Burton and Sawyer, 2016. The discussion from the Referee comment should be included in the introduction or the discussion of the manuscript.

2) The method section states that multiplications and convolutions were non-commutative (p. 4, l. 29). This is wrong, because a convolution is an integral, which is linear (see https://en.wikipedia.org/wiki/Integral). Therefore, all conclusions drawn from this hypothetical non-commutitativeness are also wrong. In DOAS, the convolution is therefore not required to be applied in every fit iteration (p. 5, l. 1). Please revise the manuscript an remove implications of this erroneous assumption.

3) The forward model (Eq. (10)) approximates broadband absorption with a polynomial factor P(\lambda) in intensity space. However, Eq. (3) suggests that it should be rather exp(P(\lambda)). What is the influence of this approximation on the fit result?

4) In my opinion, the formulation of the inversion problem is not completely clear and should be improved (Sect. 2.3.2). - Please detail, which parameters are fitted (or included in the state vector), eventually in an additional table. Please also include the initial values. - How are stretch and shift treated? They do not appear in Eq. (10).

- Is the fit applied on filtered spectra as stated in p. 4, l. 18f? - In my opinion, a real advantage of fitting in high resolution is the opportunity of retrieving an actual ILS parametrisation simultaneously. Has this option be considered? If not, why? - Figure 2 is a bit confusing. It seems that the "Final Spectrum" is entirely calculated before fitting. Please specify, which steps are done before fitting and which parameters are included in the fit. The fit is usually an iterative process, which could be sketched in the figure. - What are the termination criteria of the fit procedure?

5) The iFit results are compared to classical DOAS results in Sect. 3.3. The description of the applied DOAS method, for which the reader is referenced to Sect. 2.2 (p. 10, l. 5), is not complete. For instance, it is not clear, which software was used. From the provided description one may conclude that did not apply a commonly used and validated implementation like QDOAS. In my opinion, a standard software package should be used in order to provide a meaningful comparison between iFit and DOAS.

6) Bobrowski et al. 2010 state a "standard evaluation range of approximately 310 to 325nm". What is the effect of shifting the range for the DOAS retrieval to 305-318 as applied in this study? I would assume that interferences from O3 absorption and Rayleigh scattering are more dominating at shorter wavelengths. Please discuss this issue in the revised manuscript.

7) The results of the comparison between iFit and DOAS are compiled in Fig. 11. The discussion of the comparison, however, is a bit meagre. For example, Fig. 11 reveals a bias compared to the background (and the DOAS) value at the left and right tail. The reference was taken at 9:59, which is approximately in the centre of Fig. 11 (a). At 10:05, however, iFit results seem to be significantly positively biased. Please discuss this issue in the manuscript. Furthermore, a quantitative comparison between iFit and DOAS like compiled in Table 1 would be favourable.

8) When treating the I0 effect and strong absorption, the knowledge about the ILS is crucial. It seems that a Gaussian ILS was applied throughout the study (p. 7, l. 27). The



paper by Beilre et al., 2017 investigates parametrisations of different spectrometers and states, that the Gaussian may be a sufficient ILS parametrisation only for some instruments. Therefore, I suggest to add a plot showing the measured ILS for the applied instruments.

9) I am a bit confused about the discussion of the computational speed (Sect. 3.4). Actually, iFit requires a convolution in every fit step and, therefore, an inferior performance of iFit compared to DOAS can be anticipated a priori.

As a comparison, Beirle et al. (2013) claim to be able to process non-linear DOAS analysis using 0.004 seconds per fit with DOASIS. When they implemented their linearised method in MATLAB, they even achieved 0.00004 seconds per fit. In the study here, a faster CPU with 3.2 GHz (instead of 2.5 GHz) was applied and achieved not less than 0.09 seconds per spectrum. Hence, iFit is more than a magnitude slower than DOASIS on a slower PC.

Furthermore, the stated observation that changing the initial value could speed up the process by more than a factor of 2 indicates that either the algorithms converges very slowly or that the standard initial values are not chosen optimally. Please discuss.

In my opinion, computational speed should not be overrated for the application of a scientific algorithm. If the scientific question requires a slower data evaluation algorithm, it shall be favoured over a faster and less accurate one. The algorithm presented here works without a measured reference spectrum, which is a quality of its own. Therefore, I suggest to remove the discussion about computational speed from the manuscript altogether (also: p. 10, l. 28 "We have...")

10) I do not agree with the authors statement that fitting in intensity space is more intuitive (p. 11, l. 1). This is a personal opinion and should be omitted. For me, absorption is an asymptotic process, which is intuitively linearised by transforming from intensity space to optical density space.

* Minor comments *

- I am a bit confused about the identity of the first author. Is his first name Ben or Benjamin? Personally, I would refrain from using nicknames in scientific author lists and affiliations.

- p. 1, l. 8: "number of advantages" -> "primarily" is a bit vague. Please be more specific.

- p. 1, l. 10: remove the repeating "without the use of a reference spectrum"

- Please avoid qualitative statements like p. 1, l. 9: "accurately" p. 1, l. 10: "well suited" p. 1, l. 11: "strong potential" p. 7, l. 20: "accurately" p. 10, l. 9: "very well" p. 10, l. 24: "accurately" p. 11, l. 6: "accurately" and use quantitative statements instead.

- Sect 2.2.2 Please add a statement that the I0 effect is due to the non-commuting of exponential function and convolution. This effect can be corrected for in DOAS evaluations.

- Eq. (8): The clarity of this formula could be improved, e.g. by omitting "(\lambda)" and some more brackets. Maybe also use \frac{}{} as in Eq. (A3) in the cited Aliwell et al. 2002.

- p. 5, l. 26 omit "another"

- p. 10, l. 31: Please remove the sentence starting with "Remaining ..." because it is not based on findings in the paper. All effects mentioned can be addressed by DOAS.

- p. 11, l. 3: This is not a particular improvement of iFit, because low intensity issues are visible in raw spectra. Please omit this statement.

- Figure 4(d): Caption and legend are not matching. What is the difference between measured and synthetic Ring? Please clarify.

- Caption of Fig. 11: Please detail the definition of CST time.



- Please add gridlines to all plots to improve readability.

- Please add subplot labels like (a), (b) and so forth to Figs. 1, 6, 7, 8, and 9 for better reference.

- Please arrange the subplots in Figs. 4, 5, and 10 first left to right then top to bottom.

* References *

Beirle, S., Sihler, H., and Wagner, T.: Linearisation of the effects of spectral shift and stretch in DOAS analysis, Atmos. Meas. Tech., 6, 661-675, https://doi.org/10.5194/amt-6-661-2013, 2013.

Beirle, S., Lampel, J., Lerot, C., Sihler, H., and Wagner, T.: Parameterizing the instrumental spectral response function and its changes by a super-Gaussian and its derivatives, Atmos. Meas. Tech., 10, 581-598, https://doi.org/10.5194/amt-10-581-2017, 2017.

Burton, M. R. and Sawyer, G. M.: iFit: An intensity-based retrieval for SO2 and BrO from scattered sunlight ultraviolet volcanic plume absorption spectra, Atmos. Meas. Tech. Discuss., https://doi.org/10.5194/amt-2015-380, in review, 2016.

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