Response to Reviewer 1

We thank the reviewer for their in depth review of the manuscript which has resulted in a much improved revision. The main comment on the formulation of the forward model has been addressed with a significant re-write of the description of the fitting procedure, we hope this is now sufficiently clear. Other significant changes include a comparison with the QDOAS software instead of a custom written DOAS code and the addition of data from a scanning spectrometer station to emphasise the issue of contamination.

Major Comments

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.

The abstract, introduction and conclusion sections have been changed to reflect the advantages of fitting in high resolution rather than solely on fitting in intensity. We acknowledge that fitting in intensity is not new in of itself, however we respectively disagree with the statement that fitting in intensity is the same as fitting in optical depth due to how factors like the I_0 effect are handled.

We have included a more in-depth discussion of the corrections applied to DOAS retrievals (p. 2 l. 15 – 31).

The method section states that multiplications and convolutions were noncommutative (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.

Thank you for highlighting this fact, we have removed this from the manuscript.

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

The description of equation 10 (now equation 9 in the updated manuscript) has been expanded to explain why a polynomial had been used instead of the exponential of a polynomial (section 2.2.1).

The polynomial takes into account various factors including the Mie and Rayleigh scattering and transmission functions of the optics used.

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?

The forward model has been expanded upon (now section 2.2.3) and more detail in how the fit is achieved has been added, hopefully this is now clear. Also figure 2 has been updated to improve clarity.

Retrieval of the ILS simultaneously with the fit is possible and has been tested, although not rigorously and so was not included in the manuscript.

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.

For this comparison a DOAS script was written in Python and used to fit the spectra. This has now been changed and the comparison is now with the QDOAS software.

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 O_3 absorption and Rayleigh scattering are more dominating at shorter wavelengths. Please discuss this issue in the revised manuscript.

The lower wavelength range was chosen as it seemed to perform best for iFit, however after the comments made by both referees all analyses were repeated using the 310 – 320 nm window to be more in line with the common analysis windows. While the reported uncertainty increased (due to the reduced strength of the SO_2 spectrum) it was found that this range was just as robust as the lower window for iFit. There was also little change in the results from the previous DOAS analysis by shifting the fit window, although this has now been replaced with the QDOAS analysis.

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.

The reanalysis using QDOAS has removed this bias, and additional descriptions of the comparison have been added, including a discussion of the reported errors by both iFit and QDOAS (p. 10 l. 6 - 9).

When treating the I_0 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.

In the reanalysis a super-Gaussian is used in the stead of the Gaussian used before and this has been found to represent the measured ILS more accurately, for which we thank the reviewer. Figure 4 shows the ILS for each spectrometer has been added, and a fitted super-Gaussian function is used throughout the analysis, the parameters of which can be found in table 2. Thank you for bringing this to our attention.

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...")

This section was included after a comment from a previous reviewer of the Burton and Saywer (2016) manuscript, however we agree with the reviewer's comment and so have now moved it to the appendix. The comparison of the computation speed of iFit and DOAS will depend on how the analysis programs are written and which language they are written in. Python was chosen as it is Open Source and contains many useful libraries that can be utilised, but it can be slower than other languages. For the comparison between DOAS and iFit a similar code was written to analyse the spectra using the DOAS methodology to produce a test between the methods, not the choice of programing language.

I do not agree with the author's 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.

This comment was based on our experiences working with observatory staff, for whom the iFit program was originally designed. We agree that this is not a fact but an opinion, and so it has been removed.

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.

The birth name of the first author is Benjamin, but the name he has used in all professional correspondence, where given the choice, is Ben.

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

These statements have been removed.

p. 1, l. 10 Remove the repeating "without the use of a reference spectrum"

Removed

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.

These statements have been removed.

Sect 2.2.2 Please add a statement that the I_0 effect is due to the non-commuting of exponential function and convolution. This effect can be corrected for in DOAS evaluations.

This has been included (p. 4 l. 4)

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

Corrections made to formula (now equation 6)

p. 5, l. 26 omit "another"

This has been removed.

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.

This statement has been removed.

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.

This statement has been removed.

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

The legend has been corrected. The difference was explained in the text (p. 8 l. 5 of previous manuscript, p. 8 l. 2 in the revised manuscript).

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

CST is a time zone, this has been clarified in the figure caption (now figure 13).

Please add gridlines to all plots to improve readability.

These have been added.

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

These have been added.

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

This has been done.