

Review of *iFit: An intensity-based retrieval for
SO₂ and BrO from scattered sunlight
ultraviolet volcanic plume absorption spectra*
by M. R. Burton and G. M. Sawyer

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The manuscript *iFit: An intensity-based retrieval for SO₂ and BrO from scattered sunlight ultraviolet volcanic plume absorption spectra* by M. R. Burton and G. M. Sawyer introduces an alternative to the common 'DOAS' fit, which is applied in the spectral evaluation of stray light measurements (here: at volcanoes) in order to quantify trace gas abundances of, in this case, SO₂ and BrO.

The new approach uses a high resolution sun spectrum (also called Kurucz spectrum according to [Kurucz et al., 1984]) as its Fraunhofer reference spectrum. As the intensities are calculated at high spectral resolution, this approach does not need explicit corrections of the saturation and I_0 -effect, which are observed in DOAS evaluations due to the fact that the convolution with the instrument response function does not commute with the exponential function of the Lambert-Beer-Law [Wenig et al., 2005]. Additionally, this approach does not suffer from reference spectra which are potentially contaminated by the volcanic plume above the instrument, which might introduce a variable bias to the observations. This dependence on some 'reference spectra' is however again introduced, as later on specific residual spectra are used to correct the iFit routine.

However, the individual sensitivity of each pixel (via the flat spectrum) needs to be corrected for, as done here in the manuscript. The computational effort to perform the spectral evaluation will be larger due to the convolution operations needed to be performed within the fitting routine.

The topic presented here fits the scope of AMT and as I'm not aware of peer-reviewed publications using a similar approach I would recommend the resubmission of this manuscript after addressing a couple of individual points listed below. The new manuscript could provide information to an even wider audience, as the principle presented is not only restricted to the detection of BrO and SO₂, but could be universally applied.

In its current form, the language of this manuscript is fluent and the overall presentation is well structured. It lacks however proper mathematical formulations, carefully designed figures, intercomparison to existing methods and neglects a number of additional points to be considered (instrument line function, Ring effect, ...).

The description of calculations is not sufficiently complete and precise to allow their reproduction by fellow scientists. Furthermore the bibliography of this manuscript is quite limited and does not represent the current knowledge of related measurement principles and field observations.

1 General comments

1. It is mentioned, that the standard DOAS approach uses a number of simplifications (which are sometimes corrected for in current literature and could have been cited accordingly), such as wavelength/absorption-dependent air-mass-factors (AMFs) (e.g. [Pukite et al., 2010], an effect which is not explicitly treated in the iFit approach), assuming commutation of the convolution and exponential function (saturation effect, e.g. [Aliwell et al., 2002, Wenig et al., 2005], corrected iteratively e.g. in [Lübcke et al., 2014]), the I_0 effect [Platt et al., 1997] and more. The manuscript contains no direct comparison to the well-established, intercompared and validated DOAS approach ([Roscoe et al., 2010, Aliwell et al., 2002, Kleffmann et al., 2006, Dorn et al., 2013, Xie et al., 2004] and others). A direct comparison would be a substantial (and essential) addition to the current manuscript and could show the advantages of this new approach by comparing the results directly and discussing their differences. The DOAS analysis will be possible, as sufficient SO₂ free spectra can be found in the traverse measurements.
2. As this new fitting algorithm is based on a high resolution sun spectrum, it requires a few convolution operations of the high resolution spectra for each fit. This will slow down the evaluation process significantly compared to a typical DOAS fit and could render this algorithm unsuitable for larger datasets, e.g. data from current satellite instruments or multi-year data sets from scanning DOAS networks like FLAME or NOVAC. A more detailed discussion of this aspect giving estimates on the runtime of each of the methods would be appreciated. Even the 'normal' DOAS approach can be too slow, which is why linearisation approaches for future satellite missions (e.g. TROPOMI) have been developed, see e.g. [Beirle et al., 2013].
3. Additionally to the text description of your algorithm, a clean formal mathematical description would be helpful to unambiguously understand each step. This would also help to implement the algorithm.
4. References to possible future publications should be either shortly summarized in order to understand their possible implications or removed. From the current manuscript, it is e.g. not clear how light-dilution effects can be corrected more easily in this approach than in a standard DOAS evaluation.
5. Please try to make precise statements: The current manuscript contains a series of rough estimations and qualitative descriptions: 'We believe', 'low intensities at low SZA', 'very large numbers', 'a few spectra', 'not surprising', 'impressively small', 'relatively broad'. In my personal opinion these words seem to be unsuited for a scientific publication. If you would

precisely describe these quantities, the reader will be able to follow your argumentation more easily and compare your results to his own.

6. An important note about using a residual spectrum to correct for an insufficient spectral retrieval and to explain systematic residual structures: A correct representation of each of the effects considered in the spectral retrieval can be crucial once residual spectra are used to compensate for systematic residual structures, as the residuals linked to different effects add up in the residual used for correction. This spectrum is then scaled by one factor despite the fact that the individual effects scale independently from each other. Therefore the selection of spectra to chose the 'correction residual' from can be important for the stability of the results. This should be discussed more in depth, e.g. a time series of the OD attributed to the remaining residual structure from the fit in relation to SO₂ and BrO could be discussed. A method to extract the individually scaling contributions to the residual of the fit from a set of SO₂-free spectra can be found e.g. in [Li et al., 2013]. This point is especially important, if certain effects (e.g. the Ring effect, as shown below) are only very roughly approximated.

2 Specific comments

1. p2 line 52: Many groups refer to mini-DOAS, as they often used the 'Mini MAX-DOAS' by Hoffmann GmbH, Germany, which was named 'Mini MAX-DOAS'.
2. p3 line 65: 'from radiative transfer': The model presented in this manuscript does use a very basic radiative transfer and does not include any radiative transfer simulations. I would therefore suggest to change this phrase to 'from strongly simplified radiative transfer', or 'DOAS'-like approximation of radiative transfer, as radiative transfer effects such as Rayleigh and Mie scattering are also just effectively treated by a polynomial depending on wavelength. Thus the only difference of the iFit algorithm described in this paragraph to typical current DOAS evaluations is the use of a convoluted solar atlas as Fraunhofer reference and the calculation at high spectral resolution. This has been so far not used to calculate column densities of absorbers, but only to obtain the wavelength calibration of measured spectra (e.g. in WinDOAS, DOASIS, and other 'so called' Kurucz-fits prior to the 'real' fit to obtain the column densities of the respective absorber). Note that this approach is however used e.g. in the IR-CO₂ community for some time (see e.g. [O'Dell et al., 2012]).
3. p3 line 67 and throughout the manuscript: The DOAS method typically needs no 'clear sky spectrum' for volcanic observations, but a 'volcanic plume-free' spectrum.
4. p3 l113ff: Because there is a non-linear relation, τ is typically calculated from $-\ln(I_m/I_{sky})$ and not from I_m/I_{sky} . Then the mentioned Taylor extension is not needed at this point. The problem at optical depths of 0.1 is typically another one, and that is, that the linear operation of convolution and the exponential function do not commute (e.g.

[Wenig et al., 2005, Aliwell et al., 2002]): This leads to the so called saturation effect, which is well known and often iteratively corrected for, if necessary ([Lübcke et al., 2014]) for the relevant absorbers. For a broadband absorber, this effect might be often negligible, but especially for line absorbers (e.g. water vapour), this effect can lead to significant differences.

This effect needs to be separated from the change of effective lightpath which is induced by strong absorptions: To quantify these problems, dedicated radiative transfer calculations are needed, as e.g. the so called correlated-k method applied to absorbers such as H₂O, CH₄ and O₂ in the near IR [Buchwitz et al., 2000] for atmospheric stray light measurements. Such a model is not applied here.

5. p4 1102: The polynomial is typically not fitted separately to the actual DOAS fit, but simultaneously with all absorbers. Other filter approaches (e.g. using binomial filters of fourier transform filters) exist.
6. p6 1144ff: As pointed out above, this is often called 'Saturation effect'. Existing literature could have been cited. To circumvent calculation of high-resolution spectra to intrinsically incorporate the saturation effect, the saturation-corrected spectra are often stored in a look-up-table (e.g. [Lübcke et al., 2014]) or the effect can be often linearised within a certain range of observed slant column densities and represented by two effective convoluted cross-section or a Taylor expansion [Pukite et al., 2010]. A third method was employed in [Wagner et al., 2003], which rescales the obtained column densities obtained from a fit with a non-corrected cross-section at low resolution.
7. p7 1157ff: Here a few aspects seem to be mixed up. The iFit algorithm also does not take into account the wavelength dependence of the photon path length, as the absorption cross-section is also only multiplied by one scalar, instead of correctly simulating radiative transfer processes for each wavelength (which would require a radiative transfer model). Fitting in intensity space does not make a difference here. DOAS does not do this either, only if further corrections are applied.
8. p7 1165: iFit might require a few less manipulations, as fewer issues such as I₀ and saturation correction needs to be tackled explicitly. However, the computational effort is significantly larger. This needs to be discussed.
9. p7 1169ff: In the direct fitting approach there might not be divisions by small numbers (whatever that is), but a larger signal, from which another large signal (ozone absorption e.g.) needs to be subtracted. In terms of error propagation this will lead to the same result, as one side can be transposed to the other by a simple logarithm/exponential function.
10. p7 1178ff: As previous publications¹ the publication by [Salerno et al., 2009] contains some significant technical mistakes. It uses different instrument function widths for the convolution of different (pseudo-) absorbers in the fit. As only one instrument is used, there is only one instrument function

¹See e.g. <http://www.atmos-chem-phys-discuss.net/10/C2766/2010/acpd-10-C2766-2010.pdf>

(for a given configuration, temperature, ...). It is therefore questionable how well results can be reproduced using a correct spectral retrieval. It is however true that it uses a convoluted spectral solar atlas as reference spectrum.

11. p8 l202: The shape of the ILF is determined by the combined effects of the fibre used, the slit, the grating and imperfections of the mirrors of the spectrometer.
12. p9 l207: Why was the ILF manually determined and not fitted within iFit or separately determined using a spectrum of a line emitter? (or is it fitted, as suggested in the BrO fit settings on p24 l581 'FWHM'? This remains unclear and needs to be clarified.) How well does this parametrization represent the real instrument function? Is it kept constant for different spectrometer temperatures?
13. p9 l211ff: The readout and correction procedure for CCDs and CMOS detector signals are typically similar for various types of applications, ranging from astronomy over spectroscopy to photography. The fact that instrumental parameters change with temperature are known and published and could have been cited. Spectrometer specific properties can then still be described, if necessary.
14. p10 l231ff: As stray-light is present in virtually all spectrometers, it is typically to some amount also corrected for by a fitted intensity offset (or inverse Fraunhofer reference for a constant offset) in recent publications aiming at the detection of optical depths in the range of several 10^{-4} . This could have been cited and is ideally suited to be implemented in intensity space!
15. p10 l238: Typical DOAS applications avoid the need for a flat spectrum by dividing two measured spectra from the same instrument. In this case, it is however necessary and good that this was done and properly described. It is however no new effect and is also regularly applied in Astronomy and Photography. For hyperspectral satellite instruments this is done in the radiometric calibration step, see e.g. [Munro et al., 2015] and references therein. For GOME2 data, the pixel-to-pixel gain calibration data is obtained from LED spectra.
16. p10 l252ff: The used types of noise are not all defined, especially not 'instrumental noise', which dominates apparently the residuals. I hope this is a confusion and the residuals are dominated by photon shot noise (same error on p27 l676). From my experience various USB2000 spectrometer are only limited by shot noise at short total exposure times for RMS of the residuals of some 10^{-3} and larger, especially if large amounts of instrumental stray light are observed. Unfortunately the residuals cannot be visually examined from figures 4+8+13 due to the choice of y-axis intervals.
17. p12 l281: See comment to p3 line 65.
18. p12 l288: please cite [Kurucz et al., 1984, Chance and Kurucz, 2010] correctly.

19. p12 l297ff: 'identical', 'acceptable speed': Please quantify.
20. p13 l302 The Ring effect does not only depend on the SZA, but also strongly on the relative solar azimuth angle due to its characteristic phase function, see e.g. [Wagner et al., 2009] and references therein.
21. p13 l304ff The Ring-effect due to rotational Raman scattering (RRS) is by now well known and discussed in literature in depth: [Grainger and Ring, 1962] [Brinkmann, 1968] [Bussemer, 1993] [Fish and Jones, 1995] [Chance and Spurr, 1997] [Langford et al., 2007] and [Wagner et al., 2009] to name just a few publications show different aspects of this effect and even contributions of vibrational Raman scattering were detected in measured spectra [Lampel et al., 2015], which won't play a role here, as it leads to a red-shift of the observed sun spectrum. It is also implemented in state-of-the-art-RTM, e.g. [Rozanov and Vountas, 2014]. Therefore this effect can be effectively compensated for. Using an inverse Fraunhofer spectrum in order to compensate for the contribution of RRS in scattered light spectra was done in the 80's (e.g. [Noxon et al., 1979]), but currently these processes are better understood and can thus be better compensated for.

Using the inverse of a Fraunhofer spectrum will lead to an overestimation of the Ring effect at shorter wavelengths, as the 'redistribution' of incoming photons takes place only within a few nm, thus the absolute intensity of the Raman signal itself at shorter wavelengths is also smaller. This is shown in Figure 1. As the Ring effect is caused by inelastic scattering on N₂/O₂ molecules, most of the Ring signal originates from the lower troposphere, below the main ozone absorptions. This needs to be considered when calculating the Ring spectrum and is the reason why a measured spectrum at ground level serves often as a good approximation to calculate the Ring spectrum from.

For the publication by [Chance and Spurr, 1997] the corresponding RRS response for a high resolution solar atlas can be found and is regularly used (e.g. [Roscoe et al., 2010]). Also software packages (e.g. DOASIS, [Kraus, 2006]) include routines to calculate RRS spectra, which are basically convolutions with the spectral Raman response at the corresponding wavelength including the overall wavelength dependence of the inelastic scattering itself (for details see e.g. [Chance and Spurr, 1997] or [Lampel et al., 2015], where also illustrations can be found.).

iFit operates mostly in intensity space. Scattering due to RRS modifies intensities rather than optical depths and would be therefore suited to be directly implemented in an intensity-based fit instead of using its approximation in a DOAS fit by Taylor expansion. This is done e.g. in [Chance and Spurr, 1997].

As the Ring spectrum can, also also stated in the manuscript, contribute with several percent to the intensity of the observed spectrum, I would like to suggest to implement this effect more in detail instead of using very rough approximations.

22. p14 l346: Why is it the solar spectrum, which causes these residuals? This would have serious implications for all algorithms using these spectra

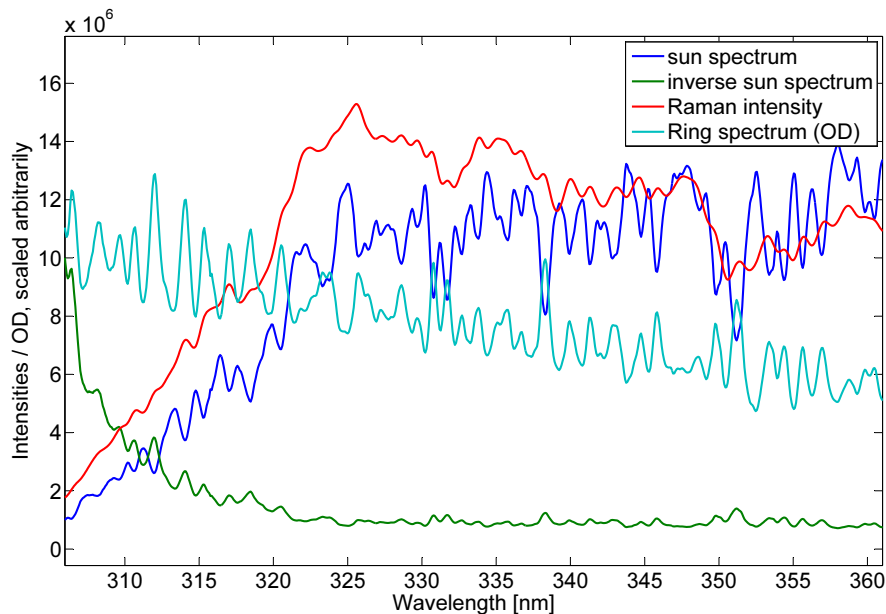


Figure 1: Overview over arbitrarily scaled spectra at 0.6nm spectral resolution of: sunlight, the corresponding Raman spectrum, an inverse sun spectrum and the Ring spectrum calculated by DOASIS.

for wavelength calibration (various satellite instruments, QDOAS calibration routine, ...). [Wagner et al., 2015] suggested a method of radiometric calibration and then compared their sun spectrum to a radiometrically calibrated spectrum. Looking at the quantum efficiency of the detector used in your spectrometer (shown in the data sheet), this could be the reason, as the total quantum efficiency of DOAS spectrometers is often not quantified. How did you exclude this?

23. p14 l348: How were these 'few spectra' selected from the observations? (Compare also: [Li et al., 2013]) In the end this selection of residual spectra is somewhat equivalent to choosing a 'clear sky reference', where is the actual advantage in this case?
24. p14 l351: How was 'pure random noise' identified? At which OD it was pure noise? When averaging pure random noise it should behave like $\sqrt{\frac{1}{n}}$, was this observed? Observations using spectra zenith sky spectra with a total exposure time of several seconds on a USB2000 can yield residual spectra of only a few 10^{-3} when evaluated against each other, why is the residual here often significantly larger? Does that show that the residual is not only 'pure random noise'?
25. p14 332ff: The ILS is parametrized by a sum of a boxcar and a Gaussian function. How was this motivated? Are real instrument functions sufficiently well represented by this parametrisation? A figure comparing a measured instrument function (from a measured spectrum of a line

source) and the corresponding parametrized representation could illustrate the differences. It is said, that the weighting and width of the two functions were 'found by inspection': How can this been done by somebody applying your iFit? Why have these few parameters not been added to the overall fitting routine? Why was not a measured instrument function used?

26. p15 l366ff: The fit presented here is most probably an over-determined system of equations, as more individual detector channels than unknowns to be determined exist. The manuscript states that for iFit an 'optimal estimation approach' is used. This is typically applied when the problem to be solved is ill-posed/underdetermined - in this case 'optimal estimation' is often applied; an a-priori is used to find a solution. Why was this applied here, what is the a-priori, which cost-functions need to be minimized by the Levenberg-Marquardt algorithm? How do the resulting averaging kernels look like? Why would a ordinary non-linear least-squares fit using Levenberg-Marquardt not be sufficient?
27. p15 l366ff: The whole paragraph lacks exact formulation of the mathematical formulae used.
28. p16 l382ff: The whole paragraph lacks citations and exact formulation of the mathematical formulae used. It could be completely removed and replaced by one citation of literature about the LM method.
29. p16 l397: A new K must be calculated, but actually is not. Please clarify and add an error estimate for the use of the empirical, simplified approach.
30. p17 l404ff: With Levenberg-Marquardt the error is typically determined from the size of the residual and the inverse of the covariance matrix of the model. Why was a 3-element boxcar smoothing applied before determining the size of the residual? This is some kind of high-pass filter - but why are broad band structures filtered out? This would neglect broad band structures present in the residual. As seen from figure 5 (which is not referenced), this leads to smaller numbers. But what is the intention of applying this filter? This could be motivated more clearly. (Again, missing formulae)
31. p18 l438ff: Without short-pass filter?
32. p19 l473f: Also the ILF changes significantly with temperature, as reported by G. Pinaridi et al in 2007 in a NOVAC report. Given the large overlaying ozone absorption in the SO₂ evaluation range, this will then still favor a temperature stabilized version of the spectrometer. Has this effect been studied, maybe together with the temperature dependence of the flat spectrum, which is shown in the manuscript?
33. p18 l438: What are 'optimal intensity levels'? Has the inherent non-linearity of the response of the spectrometer been considered (see e.g. <http://oceanoptics.com/wp-content/uploads/OOINLCorrect-Linearity-Coeff-Proc.pdf>)?

34. p24 l587: Please put these findings in relation with previous studies at Mt. Etna, e.g. [Bobrowski and Giuffrida, 2012] and references therein.
35. p24 l593: The discussion of the temperature dependent ILF is again missing, which at least to some extent renders the conclusion that not temperature stabilization is needed any more, obsolete. From my experience passive MAX-DOAS measurements during daylight are rarely dark-current limited.
36. p24 l599: This is also the case for DOAS approaches: If the broad-band part of the absorbers are not filtered out (as often the case for passive DOAS applications), the remaining residual will also provide these information. (p28 l685ff also)
 From this paragraph this argument is not clear why aerosol and ash extinction and light dilution can be efficiently and reliable implemented.
37. Figure 5: Units are missing or at least the fit error should be ppm·m?
38. Figure 6: Additional measurement errors would show if the variations from measurement to measurement are indeed correctly characterized by the measurement error. Please use the same units in all plots, if possible either ppm·m or molec cm⁻².
39. Figure 8+13: Please rescale the residual spectra to a reasonable magnitude, so that its size can be estimated directly and not indirectly from the fits of the absorbers.
40. Figure 9: The plots shown here to visualize the variation of the fit result with changing fitting wavelength interval could be seen as one-dimensional subset of the plots produced by the method introduced in [Vogel et al., 2013].
41. Figure 9+10: In figure 9 the SO₂ values decrease for decreasing start wavelength for both gas cells. Also the straylight contribution will increase, due to lower sunlight intensity. In figure 10 the values decrease during the afternoon, when the sunlight is turning more red with time. A larger relative intensity of red/infra-red light in scattered sunlight could also increase the instrumental stray-light signal, as several short-pass filter which block light in the blue and green spectral region do not block light in the near IR. Assuming the filter OF1-U325C to be a Hoya U325C filter, it is transmitting in the NIR, thus it might point towards an instrumental stray-light problem of the setup? Could a separation of the instrumental stray-light correction (e.g. constant intensity offset) and the Ring effect (using a better approximation of the real RRS signal), as done for several current trace-gas retrievals, improve the results?
42. Figure 12+14 could be combined in one plot for simplicity and readability.
43. Figure 14: The BrO slant column densities shown here were calculated from spectra recorded in the early morning of January 7th, 2007, shortly after sunrise. Looking at the magnitude of the baseline values between a SZA of ≈ 83 to 70° , these could be caused by stratospheric BrO absorption. From the manuscript it is not clear how the contribution of stratospheric BrO absorption has been corrected for. The stratospheric BrO VCD could

be determined from a Langley plots using spectra with low SO₂ values or it is found in literature. In a correlation with SO₂ dSCD this would not cancel out, as it's not constant over time.

The plot could be extended by adding the individual measurement errors as grey background, in order to visualize the relation between the variation of slant column densities within a short period of time and the measurement error.

3 Technical comments

1. Please use commonly used symbols in the equations throughout the manuscript, instead of full words. This simplifies reading the manuscript. E.g. scattering or absorption cross-section values are typically denoted by $\sigma(\lambda)$ in different fields of physics. Also please use commonly used notations for numbers, e.g. $5e^{16} \neq 5e16$, use instead 5×10^{16}
2. Please annotate each of the axis in your plots with the corresponding quantity and its unit, if possible.

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