Review of

A new simplified approach for the simultaneous SO_2 and ash retrieval in a tropospheric volcanic cloud

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General Comments

In this manuscript, the authors describe a new algorithm for retrieving SO_2 column densities, aerosol optical depths (at 550 nm) and particle effective radii from three thermal infrared (TIR) MODIS channels (bands 29, 31 and 32 located at 8.6, 11.0 and 12.0 µm). In contrast to previously described direct-fitting methods, their approach is of differential nature: For each pixel, the radiance expected in the absence of the plume is derived from the radiance measured at the plume edges. The plume transmittance is then retrieved from the ratio of the measured and expected radiances using a simplified model of the atmosphere. After this initial transmittance is obtained, a number of empirical corrections are applied. These corrections were obtained by comparing the initially retrieved transmittance with results obtained by simulating atmospheric radiative transfer using the MODTRAN model for an ensemble of geometries and atmospheric / plume conditions that are thought to be representative of the situation at Mt Etna volcano, Sicily. Finally, the authors show that when their retrieval method is applied to an example dataset, it produces similar results as other, arguably more complex retrievals relying on radiance Lookup Tables (LUTs).

The manuscript is generally well written, and although it would benefit from an additional revision focused on the use of the English language, it is fairly straightforward to follow. However, I have one major concern: The authors argue that the main benefits of this new retrieval scheme lie in its simplicity and speed (particularly the ability to do without radiative transfer modeling), thus making it very advantageous for quick data evaluation, e.g. in case of volcanic crises. For situations in which there is little or no ash in the volcanic plume, this appears to be a valid argument. Calculation of plume transmittance according to the authors' Equation 5 represents a basic (and already well established) differential absorption spectroscopy approach and should indeed be quite efficient and at least as accurate as a fitting algorithm based on LUTs.

However, the empirical corrections that need to be taken into account when plumes with any significant ash content are measured rely on the results of MODTRAN simulations for an ensemble of representative parameters. In essence, the results of these simulations themselves represent a LUT, and the correction factors are derived from this table. While it is true that using the author's approach, the LUT does not need to be published and the fit polynomial coefficients given in tables 1 and 2 suffice, these coefficients are only valid for the particular conditions (including ash type, plume altitude range, plume thickness, AOD range, sensor characteristics, mean atmospheric conditions, etc.) encountered at Mt Etna and covered in this study. Therefore, the retrieval, as it is described here appears to be specific to Mt. Etna, and cannot easily be applied to other volcanoes.

If e.g. the method were to be applied to a different volcano, a new set of coefficients would need to be calculated using a new ensemble of MODTRAN simulations. The coefficients given in tables 1 and 2 would no longer be valid. As I understand it, this is basically the same as calculating a LUT for a given measurement geometry. Since conventional retrievals that rely on LUTs are quite fast once the LUT has been generated and the measurements are simply compared to the LUT entries, it is not clear whether the described method really does simplify or accelerate the retrieval of ash optical properties in volcanic plumes, or if it only takes a slightly different route with the same level of complexity and computational requirements as the conventional retrievals.

In other words, if the correction polynomials are specific to the current measurement conditions and the MODTRAN ensemble needs to be re-modeled for other situations, then wouldn't it be easier to simply compare the MODTRAN L0 and Lp radiances (or ratios thereof) directly with the measured radiances? What is the advantage of first deriving a transmittance using a simplified model, then correcting this using the MODTRAN results? This issue needs to be clearly addressed in the manuscript, as it currently remains unclear whether this work truly represents an improvement over previously published retrieval mechanisms simply relying on LUTs.

Specific Comments

Page 8860

Line 5 – Here and throughout the manuscript, you refer to "atmospheric profiles". Please be more specific. I assume temperature and pressure profiles are needed? Anything else?

Line 5 – You state that "radiative transfer models are not required to perform the atmospheric corrections". This seems to be a false statement. You later show that you need to correct your plume transmittances using a LUT built using the MODTRAN model. Also see General Comments above.

Page 8862

Line 21 – Which volcanic ash optical properties are relevant?

Line 23 – "… make intense use of radiation transfer codes to evaluate the atmospheric corrections look up tables". Please be more specific. What type of atmospheric corrections are made? What is the real difference between the established techniques (e.g. Corradini et al 2009) and yours? Don't you also use radiative transfer models to calculate atmospheric corrections? Also see General Comments above.

Page 8863

Line 7 – Please consider renaming your retrieval. It seems as though "Volcanic Plume Removal" procedure is a good name only for the first part of your retrieval in which the background radiance L0 is estimated. After that, many other calculations are made that really don't have anything to do with removing the volcanic plume. In fact, the goal of the retrieval is not to remove the plume, but rather to

gain information on it. A suggestion for a better name might be “differential radiance” method or similar.

Line 8 – While the presented retrieval scheme does indeed calculate SO_2 and ash burdens from TIR data, it also requires a LUT of radiances simulated for the given scenario to make the appropriate corrections. Therefore, using the information contained in this manuscript, the retrieval could only be applied to Mt Etna. This sentence seems misleading and the topic should be revisited, as also suggested in the General Comments above.

Page 8864

Line 2 – Again, you state that atmospheric profiles are not needed. But they are actually used to calculate the correction factors you need later on…

Page 8865

Line 17 – Why is band 28 discussed here if it is not actually used in the retrieval?

Line 23 – If the effect of the plume edges remaining visible in the background radiance even after the plume is “removed” can be easily avoided, why do you not fix this? It would seem that if it is easy to fix, it should be done, as it will likely make the measurement more accurate, right?

Line 27 – What is the criterion that the points need to fulfill? Please be more specific.

Page 8866

Line 5 – Why is band 28 not significantly affected by the surface characteristics (e.g. temperature)?

Line 12 – You state a single variable coupling Z and T is not sufficient because of seasonal effects. Wouldn’t e.g. the geographic location and current weather conditions also affect these parameters individually?

Line 17ff – Please motivate equations 1 and 2 in the text. Besides explaining the meaning of each symbol, please state where the individual terms come from. E.g. for Equation 1, you might write “The radiance at the sensor position is given by the sum of the thermal emission from the Earth’s surface, the reflection of downwelling radiance on the surface, and upwelling radiance from TIR emission and scattering on gases and aerosols in the atmospheric column.”

Page 8867

Line 9 – You state that Eq 2 is more realistic for volcanic eruptions than diffuse degassing. While I understand the motivation for this statement, it will not always be true. Consider e.g. the eruption of Kilauea volcano (very strong plumes at very low altitudes) compared to the diffuse degassing at Mt. Etna in between periods of eruptive activity. Your model will likely describe the Etna plumes better. Please rephrase this statement to be more precise – plumes lifted to high altitudes (and cold temperatures, pristine atmospheric conditions? anything else?) will be better described by the model.

Line 21ff – The concept of a modified plume temperature is introduced here. Please give more details on how this correction is derived. Is this a standard approach also used in other models? If so, please give a reference. Currently, the given empirical terms could not be easily adapted by a reader interested in applying the retrieval to a different volcano with different atmospheric conditions.

Page 8868

Line 9 – Please explain how equation 6 was obtained. Isn't the plume emission equal to the plume absorption according to Kirchhof's Law? Doesn't this mean $\varepsilon = 1 - T_{pa}$? According to equation 3, ε would then be equal to $1-(T_p/T_{pd})$, not $T_{pd} - T_p$. I assume I am missing something here… Please clarify…

Page 8869

Line 5 – You state that the two values for T_{pdv} are empirically chosen from MODTRAN results. However, you do not explain how these were obtained. It is therefore not clear how someone wanting to apply this retrieval to a different location, volcano, and atmospheric conditions would choose these values. Or are they robust enough such that they can be used anywhere? Please provide more details on how the empirical values were obtained.

Page 8870

Line 8 – You state that neglecting the scattering term from equation 7 appears to work better when $T_{p,31}$ is larger than 0.95. This is probably true, but how do you know? Please explain. Also, you might point out that in this case, the MODTRAN correction polynomial (eq 9) is not used, so the ensemble of simulations (which essentially represents a LUT) is not needed. Under these conditions, the retrieval is essentially a conventional differential absorption approach (give some references).

Page 8871

Line 4ff – This section is a bit difficult to follow. Since you do not explicitly show the dependence of AOD (in band 32) on AOD550, it is not clear that the ratio of slopes shown in equation 12 is indeed dependent on the effective radius (although you do show this later). I suggest simplifying this a bit by referencing studies showing that the Angstorm Exponent depends on effective particle size. This proves that the ratio of AOD at two different wavelengths contains information on particle size.

Line 17 – Where is $Q_{ext}(R_e)$ taken from? Volz et al? Please cite reference.

Page 8875

Line 4ff – Again you write that only the spectral data and plume altitude and temperature are needed. But this is only true for Mt. Etna, since the correction polynomials are specific to this measurement geometry and atmospheric conditions.

Line 4-24 – Since you state earlier in the manuscript that the retrieved values depend on both the altitude and the temperature of the plume, and that the two variables cannot be combined into one because the can vary independent of one another and e.g. depend on seasonality. Why then do you

couple the two variables here in your sensitivity study? Wouldn't it be better to do two separate sensitivity studies – one for altitude and one for temperature? Which of these variables is actually better constrained? It seems that maybe the altitude itself plays less important of a role as getting the temperature right, especially for plumes in remote areas of the world.

Page 8877

Line 10 – How exactly do the LUT used in the LUT procedures (please give reference) differ from the MODTRAN simulations that were conducted in the scope of this study to derive the calibration polynomial? Aren't these very similar as well, and couldn't this be the reason why the retrieval results are so similar?

Line 26 – Again you state you do not need radiative transfer models or atmospheric profiles, and yet you need both of these to derive the correction polynomials.

Page 8878

Line 1 – Couldn't one argue that the retrieval, as it is presented here, can only be applied to Mt. Etna? If you tried to apply it anywhere else, wouldn't you need both atmospheric profiles and radiative transfer calculations to derive new correction factors?

Technical Corrections

Among other things, a number of English language corrections and language related comments are given below. Please note that this list is not comprehensive and the manuscript would benefit from an additional review focused on correct use of the English language.

Page 8859

Title: Suggest rewording title to "A new simplified approach for the simultaneous retrieval of SO_2 and ash properties in tropospheric volcanic clouds". The SO_2 and ash are in the clouds, not the retrieval.

Page 8860

Line 4 – "…only 2 input parameters…" What about the spectral radiance information? This is also needed!

Line 8 – You interpolate the "radiances surrounding the volcanic plume", not the radiances on the edge of the plume…

Line 9 – "… procedure described here…"

Line 10 – "measured BY the sensor"

Line 12 – Not sure what you mean by "highlights the plume presence".

Line 14 – Recommend either removing the word “rather” or please be more specific. How uniform?

Line 15 – Here and throughout the manuscript, recommend referring to “ocean” instead of “sea”.

Line 18 – Recommend removing “In the aforementioned bands”. This is implied.

Page 8861

Line 1 – Remove “the” before “60%” and before “80%”

Line 3 – “… recent eruptions OCCURING…”

Line 4 – Replace “the well known” with “established”

Line 5 – “… based ON look-up tables.”

Line 6 – “By recomputing the parameters of the polynomial relationship” is too vague. Readers not yet familiar with the contents of the manuscript will not understand this. Please be more specific.

Line 10 – Replace “either” with “both”

Line 13 – Remove the word “buoyant”. It does not appear to make sense here.

Line 14 – “and with THE scales and types…”

Line 16 – Remove “the” before “aviation safety”.

Line 16 – “timely ALERTS…”

Line 17 – “information are needed…”. What type of information?

Line 26 – Suggest writing “They provide the spatial distribution of a volcanic ash cloud’s total mass, mean effective…”

Page 8862

Line 3 – Remove “the” before “volcanic SO_2 emission”

Line 5 – Remove “the” before “climate”

Line 9 – “airborne and SATELLITE-BORNE”

Line 10 – Replace “provided with” with “that have”

Line 11 – “later extended to INCLUDE the 7.3…”

Line 16 – “with THE 7.3 μm…”

Line 18 – “frequently, and if the correction is not applied, the…”

Line 21 – specify which atmospheric profiles.

Line 22 – Here and throughout the manuscript, please use “plume altitude” instead of “plume height”. Plume altitude is always assumed above sea level, whereas plume height might be measured above the vent.

Line 26 – “the need FOR a quick…”

Line 27 – Why is global coverage only a potential? I thought global coverage is achieved by these sensors. Recommend removing the word “potential”

Line 28 – “derived from the multi-spectral TIR image itself…”

Line 29 – change “plume height” to “plume altitude”.

Line 29 – “temperature, while still yielding reliable results”.

Page 8863

Line 2 – “the theoretical and experimental comparisons”. Please be more specific. What are you comparing to what here?

Line 4 – “results of the CONVENTIONAL LUT retrieval approach (GIVE REFERENCE)”

Line 6 – “conclusions are SUMMARIZED.”

Line 8 – Suggest rewording : “The …procedure computes the ash AOD and effective radius as well as the SO_2 vertical column density from multi-spectral TIR images”. Also see Specific Comment above.

Line 10 – “algorithm INPUT parameters”

Line 11 – “and ash optical properties, SEE BELOW), the retrieval ONLY REQUIRES KNOWLEDGE OF the volcanic cloud altitude…”

Line 17 – “… has been adapted TO Mt. Etna…”

Line 18 – Again, please specify what you mean by “atmospheric profiles”.

Line 18 – at this point, it is unclear what you mean by “compute the MODTRAN simulations used to prepare the procedure.” Please explain what this means.

Line 19 – “the present INPUT PARAMETERIZATION of the procedure is valid only for a specific ash type (Volz et al)”.

Line 21 – Either remove “more times a day” or specifically state how often “global coverage is achieved”.

Line 23 – “atmospheric TRANSMISSION windows”

Line 24 – “radiance that reaches the sensor is PARTIALLY ABSORBED AND SCATTERED by the atmosphere”

Line 25 – Please state precisely what you mean by “atmospheric corrections”. What is being corrected? Please give references!

Line 26 – It is not clear to me why you want to “study the surface characteristics”. Do you do actually do this?

Line 27 – “remotely sensed TIR data”

Page 8864

Line 2 – replace “means no” with “eliminates the”

Line 2 – again, what “atmospheric profiles”?

Line 2 – replace “and no need” with “and THE need”

Line 4 – “during VOLCANIC CRISES when a …”

Line 5 – “are partially paid for by a decreased precision…”

Line 9 – “have been obtained on this DIFFERENTIAL basis and their great DISSEMINATION is due to their simplicity, USER-FRIENDLINESS, and HIGH SPEED.

Line 12 – “faster, and YET STILL reliable …”

Line 20 – “(AOD550) are derived;”

Line 22 – “to compute the transmittance in band 29 that is due only to the ash”. I think it would be better to speak of “radiance attenuation” here, as the ash does not actually cause the transmittance.

Line 24 – Recommend replacing the last sentence in this section with “Details of the algorithm are described in the following.”

Line 27 – “When a volcanic plume is imaged in the TIR, a dip in the radiance is typically observed along a line normal to the plume axis.”

Line 28 – Here and throughout the following text, I recommend replacing “valley” with “absorption feature”.

Page 8865

Line 2 – I’m not sure that it would ALWAYS be present (e.g. if no SO_2 and very little ash were there). This is not a necessary statement.

Line 4 – “radiance that would be detected by the sensor…”

Line 6 – “but works quite well even if… or a uniform cloud (SEE BELOW)”

Line 10 – “The radiance in the absence of the plume is obtained…”

Line 12ff – Recommend rewording to “Obtaining the background radiance by linear extrapolation of the radiance measured in the area surrounding the plume.”

Line 17 – “original MEASURED radiance…”

Line 18 – “THIS IMAGE WAS MEASURED BY MODIS Terra during…”

Line 19 – “UTC; the plume was over the ocean”

Line 20 – “the thick colored line represents the BACKGROUND RADIANCE, I.E. THE RADIANCE OBTAINED FROM EXTRAPOLATION OF THE RADIANCE IN THE AREA SURROUNDING THE PLUME.”

Line 28 – Start this paragraph with “Fig 1b shows another scan line normal to the plume in the same MODIS image. However, this time a location is chosen at which the left edge of the plume is over the ocean while…”

Page 8866

Line 2 – Remove “the” between “all” and “three”.

Line 2 – “atmospheric transmission window”

Line 7 – Replace “shortly sketched” with “schematically described”

Line 10 – “… input parameters, and these must be adjusted for every measurement to match the best estimate of the current conditions”.

Line 11 – Replace “data” with “variable”

Line 15 – “In the absence of the plume, and neglecting…”

Line 15 – Here and throughout the manuscript, you use the term “atmospheric diffusion”. I assume you mean “atmospheric scattering”. “Atmospheric diffusion” typically means the diffusion of gases in the atmosphere, i.e. it deals with the transport of gases. An example would be a plume broadening and eventually diffusing into the background air. I’m pretty sure you mean “scattering”. Please change this throughout the manuscript.

Line 16 – “radiance at the sensor POSITION is:”

Line 21 – “THIS dependence has been omitted…”

Line 22 – “In the presence of a volcanic plume, we assume…”

Page 8867

Line 1 – It is not clear to me why this section needs a new heading. In fact, the statements made in this first paragraph directly relate to the equations you just introduced in the last section. Therefore, these statements should clearly be in the same section as equations 1 and 2, and should directly follow the equations.

Line 2 – "In Eq. 2 we have: (1) neglected the increase…"

Line 2 – "because of DOWNWARD SCATTERING WITHIN THE PLUME"

Line 5 – "assumed the layer of atmosphere above the plume to be completely transparent…"

Line 13 – Since "diffusion" needs to be changed to "scattering", T_{pd} should be renamed to T_{ps} in equation 3 and throughout the manuscript.

Page 8868

Line 6 – As mentioned before, please be sure to change DIFFUSION to SCATTERING, here and throughout the manuscript.

Line 18 – "…computed using ONE OF TWO EMPIRICAL values T_{pdv}…". Also note that T_{pdv} should be remaned to T_{psv}, see above.

Page 8869

Line 1 – 6 – Please reword. This paragraph is hard to understand. Explain that $T_{psv} = 0.965$ is tried first. If T_p' is then found to be smaller than 0.75, T_{psv} is changed to 0.98. Also, "The relation $\mu = 1/\cos\vartheta$ is used to correct for the increase in path length of the slant column density when compared to the vertical column."

Line 9 – Which physical effects are not well explained by the model? I assume you are referring to the 3 effects described under point 2.3? Please insert "(see section…)"

Line 23 – The word "separately" here seems to imply that the simulations were performed separately, but this is probably not the case, or is it? Can't you use the same MODTRAN simulations and simply multiply by the two different MODIS response functions? In this case, I would omit the word "separately".

Page 8870

Line 3 – Please insert "… where a_n are the fit coefficients derived from the best fit of a third order polynomial to the plot of MODTRAN transmittance vs T_p' (see Fig 3)" after equation 9.

Line 16 – "The thick straight line REPRESENTS A SLOPE OF UNITY".

Line 17 – "The three scatter plots show a good agreement (GIVE R2 VALUES) BETWEEN THE MODTRAN PLUME TRANSMITTANCES AND THOSE DERIVED WITH OUR SIMPLIFIED MODEL."

Line 22 – "… and AOD depends on …"

Line 23 – "Fig. 4, which was obtained…"

Page 8871

Line 10 – "KNOWING m_{31}/m_{32} from …"

Line 12 – "finally the AOD550 is computed using Eq 11."

Line 16 – "is the MEAN density of the ash particles and…"

Page 8872

Line 8 – Start this sentence with "Arvani (2012) showed that…"

Line 12 – Please explain Eq 15 better: "An additional third degree polynomial was fit to…"

Line 21 – Replace "usual" with "before"

Line 22 – "and beta (..) WAS EMPIRICALLY DERIVED FROM THE MODTRAN SIMULATIONS:". Then remove the last sentence on this page.

Page 8873

Line 1 - Consider changing the title of this section to something more descriptive, e.g. "Retrieval of SO_2 and ash properties from simulated radiances"

Line 2 – Remove "theoretical part". You are testing the whole procedure here.

Line 2ff – Please change the verb tenses in this and the following section from present perfect to simple past. For example, instead of writing a comparison "has been performed", please write the comparison "was performed". Generally, "has been" becomes "was" and "have been" becomes "were". This will improve the legibility of the text and show that these actions were performed by you, not someone else before you.

Line 7 – I do not understand the sentence beginning with "This has been computed as the monthly average…". Please reword and clarify what was actually done.

Line 11 – Recommend changing this sentence to "Finally, using the described retrieval procedure, SO_2 column densities and ash properties (AOD550 and R_e) were derived for the entire set of simulated radiances and compared to the MODTRAN input parameters used to generate these radiances."

Line 17 - Consider changing the title of this section to something more descriptive, e.g. "Retrieval of SO_2 and ash properties from an example MODIS dataset and comparison to conventional retrievals"

Page 8874

Line 1 – "… characterized by A higher plume…"

Lines 1-17 – Please change verb tenses from present perfect to simple past. See comment above.

Line 26 – "the fluxes HAVE common trends."

Page 8875

Line 1 – The meaning of this last sentence of the section unclear. Please rephrase.

Line 4 – replace "characteristic" with "advantage".

Line 6 – "UNCERTANTY OF these parameters clearly AFFECTS…"

Line 8 – "SO_2 and total ash mass retrieval errors due to uncertainty in the input plume altitude"

Line 20 – remove "always"

Line 20 – Please rephrase the last sentence in this section, its meaning is currently not clear.

Page 8876

Line 2 – Begin section with "Here, the Volcanic Plume Removal procedure…" Note the recommendation above suggesting the name of the procedure is changed.

Line 4 – End sentence with "remotely sensed TIR data was described."

Line 5 – Add a ":" after "simple"

Line 6 – "independent OF the surface…"

Line 8 – "…above the plume is less negligible for low plume altitudes."

Line 12 – remove "quite"

Line 13 – "Specific TO …"

Line 14 – Suggest removing the second half of the sentence starting with "because it is…", or making this part into a separate sentence.

Line 20 – "usual TROPOSPHERIC plume"

Page 8877

Line 7-10 – please reword this sentence/ make 2 sentences out of it.

Line 12 – The sentence beginning with "In particular…" is not clear. What are you comparing to what? What do you mean by the "same differences"?

Line 14 – "… in the furthest downwind part of the plume".

Final Remarks

The authors state repeatedly that the retrieval does not require radiative transfer simulations or the input of atmospheric temperature/pressure profiles. It appears to me that this is only true for the specific case (Mt. Etna.) discussed here, and only because for this case, the radiative transfer simulations were already run for atmospheric profiles assumed to be representative of the conditions at Mt. Etna. Yet the authors also state that the retrieval can be easily applied to volcanoes worldwide. This information appears to be conflicting. As I understand it, the retrieval, as presented here, can only be applied to Mt. Etna without further radiative transfer simulations which also require the input of atmospheric pressure and temperature profiles. Only if this apparently conflicting information is dealt with in a convincing manner, and the advantages of this method over other, previously published methods can be more clearly described, can I recommend this manuscript for publication in Atmospheric Measurement Techniques.