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Atmospheric Measurement Techniques Discussions

Interactive comment on "Volcanic SO_2 and SiF_4 visualization using 2-D thermal emission spectroscopy – Part 2: Wind propagation and emission fluxes" *by* A. Krueger et al.

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Interactive Comment

Response to reviewer comments

17 October 2012

Response to referee comments on: "Volcanic SO_2 and SiF_4 visualization using 2-D thermal emission spectroscopy – Part 2: Wind propagation and emission fluxes" by A. Krueger et al.

1 General statement on the feedback of both referees

Referee comments are given black. Responses to the comments of referees are given blue.

Both reviewers agree that the work is suitable for publication in AMT, since the work addresses relevant scientific questions within the scope and presents novel concepts, ideas and tools. However, the referees criticise the way the work is written up and they ask for a revision of its presentation, including rewriting some parts. Their critical comments regarding the presentation are especially focused on sections 3.2 and 4.1, which we have rewritten in the new manuscript and the structure and order has been changed according to the comments. Also, we followed their general advice regarding notations. Both referees give several constructive comments and criticisms which are worked through in the following part.

Regarding the content of the article: Referee #1 asked for a improved discussion of errors and suggests: a) an evaluation of the measurement noise error, b) taking into account the off-diagonal elements of the averaging kernel to evaluate independence and c) to discuss if the reduction of a 3-D problem to 2-D introduces a "forward model" error or not. We realised this suggestions, however, the effort regarding the off-diagonal elements of the averaging kernel is a quite new task. No hints of how the off-axis elements can be used **without a covariance matrix** describing the variability of the interference parameters are found in the literature, neither for profile retrievals nor for the reconstruction of 2-D fields.
The problem lies in that the off-axis elements depend on the units, and only with a known covariance in the correct units the dependence between two quantities can be evaluated. The product of the off-axis element of the Averaging kernel matrix with the same element of the transposed Averaging kernel matrix is, however, unitless and might be consider as proxy of dependence. Nevertheless, the mentioned exercise and the calculated graphics are to our knowledge not "established tools" for performing a diagnostic. Developing new diagnostics would reduce understandability of the work and is out of the scope of our work. We mention that the use of model simulations might help to generate a covariance matrix and the error analysis (as suggested by Sussmann and Borsdorff, 2007) that this model studies can be applied in further work.

2 Responses to reviewer#1

2.1 General comments

The authors describe a method which uses consecutive images recorded by a scanning IR-Fourier transform spectrometer to calculate wind speed and emission rate of the volcano Popocatepetl near Mexico City. This paper is consecutive to Stremme et.al. (2012) who describe and validate the scanning IR Fourier transform spectrometer which is used to record the column density fields which are used as a base for this work. The approach is interesting and offers the possibility to measure the wind and emission strength of a volcano during day and night.

However, the paper is difficult to read and lacks important information. Sometimes the choice of terms seems questionable to me (e.g. emission flux for what I understand to be the emission rate). Because the journal is directed at a wider audience such terms should at least be defined.
We completed all suggestions regarding the choice of the terms. We now differentiate more carefully between emission flux (only twice in the article) and emission rate (normally meant). This correction was also made in the title. We added an overview page/glossary in the appendix with the used variables, definitions and abbreviations.

I therefore suggest publication but only after a major rewriting considering the issues raised below. In particular please make clear what you are actually trying to get and what you are using for it. Because it introduces a new method it should carefully be explained what you gain compared to other methods and a proper error analysis should be made. Also show the limits of the method and where to start if you want improvements.
The revised manuscript points out where the improvements are with respect to other methods (last paragraph of the introduction and a new overview section 3.1) and the limitations of the new method are stated (one paragraph in the conclusions).

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2.2 Response to specific comments

What is the main goal of this research?
We intend to present a new technique to extract information of the propagation, sources and sinks of trace gases from moving slant-column images with the main goal to determine the emission rate more accurately.
Getting the wind speed or the amount of the emitted gases from the volcano? As the title states, the main goal of this research is to provide continuous calculations of the emissions rates of gases, but the wind-speed is part of the result and a quantity which can be compared for validation purposes with model data or radiosoundings.

2.3

The authors describe at length the retrieval of the wind speed. The calculation of the emission flux (meaning the amount of the emitted gases?) is only described in the section 4, labeled "Diagnostics and errors". But it seems to me, that at least one main result of the paper, the calculation of the amount of emitted gases is this calculation.
We have now separated the sections "The calculation of the emission flux" from the "Diagnostic and errors" in the revised manuscript and moved them to the methodology section.

2.4

The authors often mention the wind speed, which is retrieved. But the method is only suitable to retrieve the component of the wind speed which is perpendicular to the line of sight. Please clarify this throughout the whole article.
This is correct. It was originally stated but we now emphasize it and mention it always when we refer to the velocity. The chosen variable v_{2d} refers to this fact throughout the

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whole article.

2.5 Error due to wind direction

How has this been dealt with in the calculation of the emission flux? The authors give an error for the miscalculation of the distance of the plume in section 4.4.1. They do not mention the direction of the wind. If it does not matter, please state so and justify. I consider this crucial, because the method is not very useful if the wind direction cannot be dealt with appropriately.
Yes, indeed the error due to a wind component in the direction of the line-of-sight, does not directly affect the calculated emission rate. The product between the slant column and the wind component perpendicular to the LoS stay somehow constant. If your view is not perpendicular to a plume, the measured slant column will increase with 1/cos(α), but the wind component you see decreases by a factor of cos(α), alpha is the deviation of the viewing angle to the angle perpendicular to the main direction in which the plume propagates. An example: If the wind is 45° to the line of sight. How does it influence the result of the emission flux? The example really makes it clear and we have added this one and the explanation above. How large is the error. How do I decide if there is no wind, weak wind or the wind direction is more or less parallel to the line of sight. As shown above, the problem is the displacement towards the observer, which introduces an error in the conversion from changing angle to the "real" velocity. A possibility that the plume width increases greatly with time also exists and this limits the evaluation when the wind direction is not perpendicular enough. Many criteria are not well determined like the chosen frame rate, frame and step sizes, and will depend on the current propagation speeds, emission strength and wanted accuracy. Especially the latter is an important criteria which has to be evaluated according to the objective of the study.

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2.6 Three steps

The method seems to consist of three steps, a retrieval of the wind field, a measurement of the wind strengths and a final retrieval which uses the results of the first steps to retrieve the source distribution. I found it rather difficult to find that out, especially because the second and third step is described in what appears to be the error section. I would suggest an overview of which steps are calculated at which time using which previous results and which constraints is provided in section 3.1. Also an overview of the determination of the constraints, which sometimes uses results of one of the steps would be very helpful. We agree and present now a section and a table which gives an overview of the proceedure and complete retrieval scheme.

2.7 Rudimentary error discussion

The error discussion is very rudimentary. No statement is made on the error of the wind speed and wind direction. The conclusion of the error on the emission flux can only be believed but it is not conclusively derived. We have added a section called "Total error" (Sect.4.3.5. of the revised manuscript, page 16, line 495) with proper discussion, the resulting overall error is around 35%.

2.8 On the independence of the source strength and the wind field

The statement regarding the independence of the source strength and the wind field is at least questionable. In any case, it has to be proven, that this statement is valid. We corrected the statement on the independence of source strength and wind-field and added a small discussion about the interference error.

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2.9 Order of pictures

It is common that the pictures are in the order of their first reference in the text (Page 8, but more places). I would recommend to stick to this custom, because it makes reading more easy. Corrected. We changed the order of figures according to the custom. Former Fig. 7 is now Fig. 4.

2.10 Projected wind speed

The fact that the method can only retrieve the component of the wind speed projected to the plane perpendicular to the line of sight should already mentioned here, not partially and every now and then in the paper.
Done, v_{2d} has now a subscript to always remember this fact.
Equation 2 and 3 can only be regarded equal if the velocity has been projected to the plane perpendicular to the line of sight (LoS).Eq. 3.2. Correct. The way how the projection is realised is due to the so called "Curtis-Godson approximation", therefore, the results for the emission rate is correct even when the velovities in the wind-fieldare not homogeneous in the z direction. (Eq. (3) page 5., revised manuscript). However, the "projected" wind velocity (averaged velocity) is only similar to the real wind velocity components in x and z direction if the velocity field is more or less homogeneous.

2.11 Notation scalar-field, vector-field

As I understand the principle so far, cl is a scalar field and v is a vector field. The style of the variables in Eq. 3.2. should show this difference (please refer to the guide lines of the AMT journal on how to do that). Done and also a table (Appendix C) with the used abbreviations and definition of variables is included in the revised manuscript.

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2.12 Forward model

The calculation of the forward model seems rather strange to me: How is the gradient of the column density distribution been calculated, from the difference of the two consecutive images which also forms the measurement (dcl)? If this is the case I wonder if this is circular. Correct, some information was missing: We added (revised manuscript page 6. Eq.(7) and (8)) the gradient of the column density field is calculated from the average of two consecutive images. Two equations to explicitly present dcl/dt and grad cl have been added. The gradient of the column can be seen in the images by the change in the color and is similar to the Averaging kernel for (x or y).

It seems to me that either the wind field or the source distribution and strength must be known in order to get the other one. Please explain this in detail. No, both can be retrieved simultaneously. How well this is done depends on the measurements. We added a sentence to address this question at the end of the introduction as it is something new.

Please state the structure of the solution vector already at this place. It makes it really difficult and confusing to get this information later in the paper. Done (revised manuscript page 6, line 162).

In this section two different symbols for div (sometimes also ∇) and grad (sometimes ∇) are used. Please use one of those and stick to it consistently throughout the paper. We removed the equation using operators for the whole field and use the tensor notation, component wise (revised manuscript page 6 Eq. (11), (12) and (13).

Equation 5 and 6 What is x and y? The axis of the plane in which the wind retrieval takes place? If so, please state it. Done.

What is the partial derivative of x and why is it necessary? It is a component of the gradient. The forward model needs it. What is cl in equation 6? The column density

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field of the first or the second image, or a mean thereof? Correct, it is "the mean of them", as we already stated in the question above we make it explicit in the revised manuscript Eq. (7),(8). Why is the divergence given as an differential operator in equation 6 but not the gradient? This point addresses actually the general criticism of both reviewers, even they finally understand the work. "$grad_x cl$" is explicitly shown, the gardient of the column density filed is already calculated, the "div_x"-operator has still to do its work on the 2D velocity field (or more exact on the x- component of the field). Reviewer 3 suggested to expand Div (v*cl)=(grad cl)+cl (div v) in the Eq. 3 (amtd manuscript) . (see the expand equation in the revised manuscript page. Eq. (5))
How is the gradient calculated if not from two consecutive images? It is a gradient in space from neighbouring pixels, actually it is calculated from the average of the two images, but it can be calculated for each image.

2.13 Inversion

The description of the retrieval is not very clear. The authors mix the concepts Optimal estimation and Thikonov regularisation in a very arbitrary way and use terms in their own fashion.

I would strongly recommend to stick to the usual meaning of the terms and clarify the description of the retrieval. Examples: Thikonov-type smoothing constraint: Thikonov describes a method to solve an ill-posed problem using a weighted mean of the data norm and some constraint. In the more general Thikonov regularisation the form of the data norm and the constraint is left free as long as certain conditions are fulfilled. As I understand the authors mean the L1 norm, which they also state later in the paper. Correctly, we avoided L_1 as it was used as layer 1 in Part 1 (Stremme,2012), but we decided that it is anyway better to include L_1 in the revised manuscript.
The Bayesian approach means that the ill-posed problem is tackled using Bayes theorem. A priori information is always used, also in the Thikonov regularisation, because

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one uses information which is known before the measurement took place (knowledge a priori) and is not restricted to the Bayesian approach. It only takes a certain form in the Optimal estimation method described by Rodgers (2000) and this is mainly because of tractability. Please sort out your use of terms and concepts. A good starting point to do this is Rodgers (2000). We rewrote parts of the section and use L_1 operator instead of D_1 (despite tha L_1 was used for the layer 1 in the radiative model description in Stremme 2012 (Part 1) and x instead of $\mathbf{V}$. There shouldn't be now a conflict with the definitions in Rodgers (2000) Page 9 line 1. I do not understand how the residual shows the impact of the chosen weighting. Is the residual calculated before or after the retrieval? The residual is calculated with the retrieval and therefore afterwards. It is the difference of the simulation (using the retrieval) and the measurement. A large residual is expected where the retrieval deweights the measurement $(\mathbf{S}_e^{-1})_{l(i,j),l(i,j)} = 0)$. The effect of systematic deweighting is also described as a strategy to reduce the H_2O interference on the SO_2 retrieval in Part 1 of Stremme et al. (2012).

Page 9 lines 3ff. A few lines above the inverse $S1_e$ is calculated from the residual, now some correlation coefficient comes in. The correlation coefficient is a quantity calculated from two random variables. What are those variables and what is correlated here?
The variables are the measured (a) and reference spectra (b). See Part 1. Stremme et al. (2012) Why is the root mean square of the fit linear to (1-correlation)?
$(r^2 = 1/N \cdot \sum_i^N (a_i - \hat{a}) \cdot (b_i - \hat{b})$,
The residual (RMS) is given by $RMS = 1/N \cdot \sqrt{\left(\sum_i^N (a_i - b_i)^2\right)}$ Please redo this section and make clear what you are actually doing. Done, we describe the initialization of the $S1_e$ diagonal matrix in more detail in the revised manuscript (Page 7 line 188-222). The equation how it is calculated is given in line 200. $\left(\frac{(r^2_{Pearson} - r^2_{threshold}}{r^2_{max} - r^2_{threshold}}\right)$

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2.14 Sections

I guess section 3.3.1 is the following section. Please change your numbering accordingly. Done.

Page 12 line 21. Which equation is meant here? We have added here "terms 1 and 2" of equation 14 (AMTD) and 15 (revised manuscript, page 7).

Page 13 line 12. What is meant by adequat? How is the smoothing determined, manually, automatically? Adequate. The smoothing is adjusted manually and empirically by the operator.

Which equation is meant here? done, we add to "terms 1 and 2" of equation 14 (AMTD) and 15 (in the revised manuscript, page 7).

Page 13 line 12. What is meant by adequat? How is the smoothing determined, manually, automatically? Adequate. The smoothing is adjusted manually and empirically by the operator.

Which series are cross-correlated and to what purpose? Please make this clear in the beginning of this section. We start the section with following sentence: "The emission rates time series calculated with the columnar distribution of the first image and the retrieved wind-field and the emission rates time series calculated with the columnar distribution of the second image and the retrieved wind-field". It should be clear in the revised manuscript (Sect. 3.5 Page 9 line 264 ff.)

Page 14 lines 11-12 Using the method described above? Other method? Please provide examples of how the new constraint changes the retrieved wind field. The wind speed after the first retrieval is sometimes wrong (revised manuscript page 9 line 261). The final retrieval uses the improved *a priori* wind velocity which allows for an optimal estimation type retrieval. This damps the difference between the mean velocity and the mean velocity of the *a priori*, but it has a reduced smoothness effect. The final result shows both a correct mean wind velocity and variance of the wind velocity in the field.

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Equation 15, 16 Define latter and former. cl are scalars and v and j are vectors, please change your notation appropriately. Done.

Page 14 line 17. Which trajectory?
The main trajectory of the plume indicated as solid line in Figure 3.

2.15 Emission flux definition

Page 15 line 1. Emission flux is defined here for the first time. Please define this variable in the beginning of your paper, because it seems one of the major outcomes of this work. If not, why is it calculated? Page 15 lines 14,15. We defined "emission rate" at the end of section 3.1 of the revised manuscript, p. 4 l.115. We corrected the use of emission flux and emission rate.

2.16 tshift, tframe

What exactly is tshift and what tframes. I never found this variables again. The variables are needed for equation 24, t_{shift} is representative as the location of the maximum in Fig.5 and t_{frame} as the vertical line in the same figure. We added this statement in the revised manuscript.

2.17 AVK and its off-axis elements: Section 4.2

Are the AVK only calculated from the third retrieval? If so, the statements made below, that the retrieval of the wind speed in x and y direction and sources are independent is wrong. The statement was badly formulated and ambiguous: "The DOFx, DOFy and DOFsrc **are** independent quantities and have the values 68.6, 92.7, and 11.7,

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respectively". The sentence was changed to: "The DOFx, DOFy and DOFsrc **describe** the independent quantities (Rodgers, 2000) and have the values 68.6, 92.7, and 11.7, respectively. In addition, we added the citation to the textbook (Rodgers, 2000).

Even if they would be so, please do actually show that the results for those three components are independent using the off-diagonal values of the AVK, which are calculated for all steps. This can be used be a perturbation calculation if an analytic expression cannot be found. The reviewer addresses here the so called "interference error", an error which is missing in the error description by Rodgers according to Sussmann and Borsdorff, (2006). The problem is that it depends on one hand on the *a priori* and on the other hand on the true state of the atmosphere. For a unconstrained retrieval this error will be almost zero, if there are no other errors in the forward model. However, stronger constrains result in an increase of all random errors (measurement noise errors). As we stated at the beginning of this document, the error can only be evaluated with a known covariance matrix of the source flux and wind components. We calculated the full averaging kernel, but we did not find a proper and easy way to use its information. We think that developing new diagnostic tools in this direction might be interesting but out of the scope of this work. The result for the distribution of the source flux field seems plausible and a correct simulated source flux does not produce an interference error and the error due to interference might be limited.

2.18 Forward model error and parameter error: Section 4.4

Rodgers (2000) distinguishes between forward model error and forward model parameter error. Described here are the errors in the forward model parameters, not in the forward model itself. Correctly and to stay consistent with Rodgers(2000) we changed the section to "errors regarding the forward model" and address this definition in the revised manuscript in the lines 440 page 14.

Page 20 line 2. Please cite properly which Part 1 you mean. Done. Page 20 line 9.

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What is meant by the velocity of the viewing angle dϕ/dt ? Please define. We changed the formulation in the revised manuscript (p. 14 l. 430) to "In fact, only the change of the angle in which the plume appears $\frac{\mathrm{d}\phi}{\mathrm{d}t}$ can be obtained from the measurement".

2.19 Measurement noise matrix: Section 4.4.2

If it is not possible to calculate a proper measurement noise matrix, you should provide an example using artificially created fields of column densities given a wind field and a source distribution. Done. We have basically rewritten the section (revised manuscript p.15 l. 448-461). The noise of 13% resuts in an error of 20%. We thank the referee especially for the suggestion in this comment.

2.20 Smoothing error

Page 21 line 4. Rodgers (2000) defines it differently, pointing out that his first definition was wrong. In particular, the smoothing error is only accessible if the statistics of the original is known.
The reviewer refers to the definition introduced by Rodgers(1990) of "smoothing error" as it is normally used and applied also here. We actually discuss explicitly both, the satistically based smoothing error (eq.21) and the phenomenon of the smoothing of one particular profile. We agree with the reviewer, but we do not see the point to explicitly call the definition in Rodgers 2000 "wrong" and we leave it like is.

Page 22 line 9. Perpendicular to what? We added "to the line-of-sight"
Page 23 line 9. Please cite the paper which is part I. Done.
Figure 1. Which time is needed to take one image? Can they be considered as snapshots or are they taken continuously? The instrument is based on a single detector and a scanning device thus the measurements are continuous and the images take some

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time to complete. New hyperspectral devices could improve this. We added that in the figure caption and state it in the last paragraph of the conclusions.
Figure 3. The "drawn trajectory" is almost invisible. Please make it stronger and put an arrow at it. Done.

3 Anonymous Referee 3

3.1 General comment

The paper by Krueger et al. reports on a method how to estimate the source strength of a volcano from consecutive measurements of the 2D gas distribution in the volcanic plume. The method simultaneously derives information on the winds driving the plume. Krueger et al. apply the tool to mainly the SO2 concentration field observed by thermal emission spectroscopy of the Popocatepetl volcanic plume as described in a precursor paper [Stremme et al., 2012].

The method - exploiting the continuity equation - is appealing because of its conceptual simplicity and its ability to estimate the winds simultaneously with the source strength. One might think of applying such a method to other source/sink questions in atmospheric sciences, e.g. to be addressed through geostationary satellites. The topic of the paper is well suited for publication in AMT.

The major shortcoming of the paper is the way how it is written up. The paper requires a major revision with some parts probably requiring a complete rewriting. My comments below point out some of the major issues but the whole paper is to be examined keeping in mind that the educated reader should be able to follow the rationale with adequate effort. I skip numerous issues concerning wording and typos.

3.2 Section 3.1:

*The physical meaning of equation (3) is to be described in more detail. If I understand correctly: We added two equation (2), (3) in the revised manuscript (p.10) and expanded Eq. (5) revised manuscript (p.10).(former Eq (3)). - the 3rd dimension (along the line-of-sight) is neglected in a sense that the 2-D wind speed vector is independent

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of the line-of-sight.
Correctly it is not neglected, but it is averaged by integration. We include the calculation in the appendix (A) of the revised manuscript. For the emission rate it does not matter if the 2-D wind-field vectors change along the line-of-sight.
The z-component is the direction of the line of sight is also eliminated by integration and not neglected. $\partial_z \cdot \int_{instrument}^{horizont} v_z \rho dz = v_z(horizont) * \rho_{horizont} - v_z(instrument) * \rho_{instrument}$. This term disappears completely, for the molecules with a low atmospheric background like (SO2,SiF4) $\rho_{horizont} = \rho_{instrument} = 0$. However this argument does not apply automatically for other applications and is actually the term which allows for a top down emission estimation based on solar absorption (Stremme,2012b). Actually even if the plume would leave the field of view in the direction of the instrument, the missing term would be compensated by a source term.

- the source term Q is not the true source strength of the volcano [units: mass/time(/area of the volcano)] but it is the source integrated along the line-of-sight [units: number of molecules/area perpendicular to the line-of-sight/time].
The term describes local source flux integrated along the line of sight. We added in p.5 l. 143:
" The source term has the unit of molecules per area per time, which is equivalent to emission flux. The term accounts also for chemical processes as e.g. SO_2 destruction through photolysis or dissolution into the aqueous phase." - the source term Q is estimated from the measured slant column densities. So, Q will also depend on retrieval sensitivity along the line-of-sight.
The reviewer is correct:
If the retrieval has different sensitivities in foreground region the plume region and the background region and there is a flow along the line of sight, a erroneous sink or source will be retrieved. Estimation and quantification of this error would need to use an averaging kernel for slant column retrieval along the line of sight. Actually this averaging kernel is not available and should first be addressed and implemented in the algorithm

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described in Part 1 (Stremme et al., 2012).
- chemical processing in the plume is neglected. This assumption should be justified based on some simple photochemical considerations.
Actually they are not neglected. It is part of the retrieved source strength. It is explicitly stated in the revised manuscript (page 5, line 140-144) . (Maybe it can be suppressed in some sense due to the choice of the *a priori* information and the constraint). Figure 7 (Fig. 2 in revised manuscript) shows a sink in the center of the plume with some distance to the crater. However for the use of this retrieved information about photochemical destruction and the lifetime of SO2, first the mentioned dependence of the retrieval sensitivity along the line of sight should be studied in more detail, before conclusions based on the retrieved source distribution are made.)

*The choice of "boldface(cl)" as a symbol for a scalar quantity is very confusing. Why not just "c"?
We use "cl" without boldface in the revised manuscript c remembers easily to concentrations.

It could be helpful to expand "Nabla(v*cl)" (equation 3), because this is what the reader needs to understand equations (4), (6), (7).
done. (Eq.5 , revised manuscript, page 5 line 145,)

$$\begin{aligned}\frac{\mathrm{d}cl}{\mathrm{d}t} &= -\nabla\cdot(\vec{v_{2d}}\cdot cl)+q \\ &= -(\nabla\cdot cl)\cdot\vec{v_{2d}} - cl\cdot(\nabla\cdot\vec{v_{2d}})+q \\ &= (-[\underbrace{\nabla\cdot cl}_{term\ 1}+\underbrace{cl\cdot\nabla}_{term\ 2}],\mathbf{1})^t\cdot(\vec{v}_{2d},q)\end{aligned}$$

*It would certainly be helpful to explicitly introduce the discretization step "dcl -> clt+1 – clt" at the end of section 3.1 with t and t+1 referring to the observation at time step t

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and t+1.
done. (Eq. (7), revised manuscript, page 6)

$$\frac{\mathrm{d}cl}{\mathrm{d}t}(t_n) = \frac{cl^{t_{n+1}} - cl^{t_n}}{t_{frame}} \quad \text{here}: \ \mathrm{d}cl := cl^{t_{n+1}} - cl^{t_n}, \ \mathrm{d}t := t_{\mathrm{frame}}$$

3.3 Section 3.2:

*Choosing the state vector "capital, boldface V" is confusing because capital boldface symbols typically refer to matrices. The letter V further hints at wind speeds (v) but there is also source terms in the vector V.
Done, We changed "capital, boldface V" to x, so as the atmospheric state vector in (Rodgers, 2000)
*It is very difficult to understand the rationale in this section with the state vector being introduced at the beginning and its components being listed at the very end.
Changed, the vector is introduced just before it is used in equation (9) revised manuscript and its components are described within the next 3 lines. (Page 6 around line 160, revised manuscript)
*If I understand correctly, the state vector contains the wind speeds and the source terms for each pixel (i,j) of the 2-D image of the observed scene. The 2D scene is made a 1D state vector by chaining the pixels (i,j) into a column vector (while keeping the double indices). This process is to be described at the beginning of the section.
It is now described at the beginning of the section (revised manuscript p.6, l.162). And is more explicitly described by the relation between the indices $l(i,j) = i + (j-1) \cdot m$ (revised manuscript p.6 l.172).
Presently, the derivation of equation (6) and (7) is not understandable. *I do not understand the representation of "divmn x " in equation (6). *Would it be useful to write out equations (4) through (8) for one of its components "dclij=: : :"?

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Yes, We removed equation 6,7 and give the equation in the notation of tensors or for each element.
(Equation 11-13, revised manuscript page 6)

$$(\mathbf{K}_x)_{l_{(i,j)},k_{(i',j')}} = \underbrace{\left(\frac{\partial cl}{\partial x}\right)_{l(i,j)} \cdot \delta_{lk}}_{\text{from term1 in Eq.5}} - \underbrace{cl_{l_{(i,j)}} \cdot \delta(j,j') \cdot \frac{\delta(i,i'+1) - \delta(i,i'-1)}{2\Delta x}}_{\text{from term2 in Eq.5}}$$

3.4 Section 3.3:

*There is reference to section 3.3.1, but it does not exist.
Corrected to "next paragraph" (Page 7, Line 204 in revised manuscript)

3.5 Section 4.1:

*Section 4.1 introduces the “cross-correlation” method. To a large extent, I was not able to follow the rationale in this section. I was not able to understand why you need the cross correlation method. Do you need it for an a priori estimate of the winds (p.4613,l.17)? Please rewrite this section:
We rewrote the section and included it in the "methodology" section. Why “cross-correlation” is needed we describe in the new overview section (Sect.3.1, revised manuscript, p. 4)
- I suggest starting with a clear outline why you need the cross correlation method in addition to the method presented earlier.
done, we introduced a overview section. (please see response to former comment.)

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Interactive Discussion
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3.5.1 - What is the first retrieval you describe in paragraph p.4612,l.7?

The sentence was corrupt. It is changed to: "First, the 2-D wind-field is retrieved using a pure smoothing constraint and the spatially average wind velocities are calculated (for the average the wind is weighted by the SO_2 column). " Is it a cross correlation retrieval or the full retrieval? If it is the latter, it seems to be modified from the setup the described in section 3. That is very confusing.

Cross correlation is not a retrieval, it is more a calculation. The cross correlation as in the cited literature correlates two time series. two obtain two time series the information of the wind velocities are needed and a first a retrieval of the 2-D wind-field is necessary. This first retrieval is characterised by its "constraint" as smoothness constraint optimized for the wind direction. There are no cross-correlation-retrieval. Cross correlation just reports the time lag (t_{shift}), which results in the largest correlation coefficient. It should be clear after introduction of the overview-section (Sect.3.1, revised manuscript,Page 4) and a overview table (Table 1 revised manuscript).

3.5.2 - paragraph p.4612,l.13: Suddenly, the images are treated separately.

Do you start describing the cross-correlation method? If so, it would be good to add text that summarizes how the cross correlation method works conceptually.

Done." Usually two time series measured at different location are used for the cross-correlation. In this work two time series are reconstructed with the help of a 2-D wind-field from spatial distribution recorded at two times." (Revised manuscript page 10 line).

Revised manuscript page 10 line 274-291 describes how the emission rate time series are calculated. The use of the time-lag (t_{shift}) resulting from the cross-correlation is described by the equation (23).

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3.5.3 - p.4612,l.17: What is the “final (third) retrieval”?

There is no hint in the manuscript that actually three retrievals are performed.
This should be clear from the "Overview section", (line. 112 page 4, revised manuscript) and table 1

3.6 Section 4.2:

*The use of “boldface(AKijkl)” for the averaging kernel matrix is again confusing because the notation looks like a matrix multiplication of A and K.
We use $\mathbf{A}$ in the revised manuscript.

*p.4616,l.8: Is it really necessary to introduce “AK_θ” and “AK_ϕ”? The discussion would not suffer from just using “AK_x” and “AK_y”.
We agree and we removed the “AK_θ” and “AK_ϕ” section.

*p.4616,l.21: Is there really non-zero averaging kernel diagonal elements in the vicinity of the volcano (lower panel of Fig. 6)? I cannot see it. Consider changing the color scale of Fig. 6.
We included in the revised manuscript also the averaging kernel of the first retrieval and this shows a slightly stronger sensitivity. Therefore it is not necessary to change the color scale.

3.7 Section 4.4.1:

*Why do you need to estimate the distance r between the observer and the plume? Estimating r seems difficult because the method uses column densities integrated along

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the line-of-sight.
Is it because you want to translate radial velocities (units angle/time) into Cartesian velocities (unit length/time)? Why do you need Cartesian velocities at all?
At first the distance is necessary to determine the amount of gas and the emission rate. Even if column density is already integrated in one direction the result has to be multiplied with an area to calculate the amount of gas in the image or with an line and a velocity for the emission rate. The wind speed in Cartesian velocities is a quantity, which can be compared and validated against e.g. radiosondes,results from models.

At first approximation r can be easily estimated as the distance to the crater (12 km). For the mentioned example applications using the method with geostationary satellite it would be 35,786 km and indeed the relative error would be neglected against all other error sources and for another example the use of the method in Krueger (2012a) also to characterize horizontal trace gas flow based on measurements of an imaging DOAS device. The boundary layer height based on ceilometer measurements was used.

Nevertheless we agree with the reviewer in principle, that it would be more correct to stay by the original geometry and looking for a way how to retrieve the plume distance and improve the method to handle the 3^{rd} dimension, but it will be more complicated to interpret these data. We think that it is better to limit the discussion to cases of almost perpendicular plume propagation and to estimate the distance by the distance between the measurement site and the volcano.)

3.8 Section 5.2:

*The paper gives emission estimates in units mass/time but the retrieval method yields emissions integrated along the line-of-sight (number of molecules/area perpendicular to the line-of-sight/time). What assumptions go into the conversion e.g. relevant length of the line of sight/extent of the plume along the line-of-sight?
There are not too many assumptions but it is mainly the usage of Gauss's Law (see

revised manuscript, Page 10 line 274-284).
The only assumption is that there is only trace gas in the defined plume and the "closed surface" around the Volcano, has to be chosen (virtually) so that its intersection with the plume is completely inside the area of the image- The line-of-sight has to be in the chosen surface, an example for the other is shown as a line perpendicular to the main wind direction in Fig.3. The area in which the plume intersects the chosen closed surface is given by the one line in the image and the z dimension in direction of the line of sight. $\vec{F} = \rho\vec{v}$, $Q = \int_{Area} \vec{F} d\vec{A} = \int_{rectangle} \int_{LoS} \rho\vec{v} dz dy$ with $d\vec{A} = dz \times dy$

To calculate the emission rate from wind velocities and column measurements, integration over a line is needed. This line should start and end outside the plume, but it should cross the plume. Favourable the crossing-line should be perpendicular to the plume propagation, but it does not matter if the line are not exact perpendicular. (As the line-of-sight is in the plane the normal vector of the area is perpendicular to the missing dimension. Movements of the plume in the direction of the LoS does not affect the calculated emission rate. The only reason to chose an almost perpendicular cross section is that it could be assumed that the gas along the line was emitted at the same time.)

*Fig. 1: Color scale missing. At least give a hint on “warm” colors meaning high SO2, “cold” colors meaning low SO2 concentrations. done Why does the figure caption say “the average wind vectors are sought”? My understanding is that the proposed method tries to estimate the wind vectors in all pixels.
It should be a general motivation, telling the question what is thought to be answered with this work. We changed to:" The wind vectors describing the plume propagation projected in the image-plane are sought."

*Fig. 2: It might be useful to show the position of the volcano.
Done for the revised manuscript, we indicate the position of the volcano as a black solid line. I do not understand the last sentence of the caption.

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No wind-field can be reconstructed if no trace gas is detected, that is why the retrieval deweights the regions in which no trace gas is determined or the retrieved slant columns are classified as "not reliable" , the residual outside the plume is larger. "is almost zero at the plume position as the algorithm weights the fit with $\mathbf{S}_e^{-1}$. " was changed to: "Due to the choice of $\mathbf{S}_\mathrm{e}^{-1}$ (see text, Sect. 3.4), the residual of the column differences are larger outside of the plume."

*Fig. 7: “SAinv Sources” has never been defined or discussed in the manuscript.
We corrected the title to: Diagonal of constraint regarding the source flux
The unit of the sources should be “per time”.
"Source are changed to "reconstructed source flux" and the unit is corrected to molec/(cm^2 s)

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