

Interactive comment on “Retrieval of absolute SO₂ column amounts from scattered-light spectra – Implications for the evaluation of data from automated DOAS Networks” by Peter Lübcke et al.

Anonymous Referee #3

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Review of Manuscript AMT-2016-24: Retrieval of absolute SO₂ column amounts from scattered-light spectra - Implications for the evaluation of data from automated DOAS Networks by P. Lübcke et al.,

Recommendations I recommend publication in AMT only after a MAJOR REVISION of the manuscript in line with criteria requirements of the Journal.

General comments This paper discuss the implementation of a modelled Fraunhofer Reference Spectrum (FRS) for retrieving SO₂ Column Density (CD) from open-path ultraviolet spectra by scanning ultraviolet spectrometer. The authors carry out a number of specific statistical analysis (e.g., PCA) such as to identified instrumental features of spectrometers and explore confidence in the retrieval. Data from NOVAC network at

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Nevado del Ruiz (Colombia) and Tungurahua (Ecuador) between 2010 and 2012, and 2009 and 2011, respectively were reduced applying both the standard NOVAC (e.g., Galle et al., 2010) and the FRS approach. Comparing the results gathered from both methods, the authors observed a large difference between the two, with the NOVAC underestimating SO₂ CDs more than a factor of two. At volcanology observatories self-reliance from robotic system is a first priority for real-time multidisciplinary surveillance and monitoring purpose. NOVAC is largely spread at active volcanoes worldwide representing a unique trail for the understanding of volcanoes and their gas emission impact in climatology. Hence, the application of the modelled FRS at NOVAC represents a step forward for both monitoring purpose and revaluation of global volcanic emission rates inventory.

I believe this paper is in line with the scope of AMT. However, there are some aspects of the work that are questionable, and in which a strong revision is required before the manuscript is suitability for publication in AMT. There are also several minor points to address, which I have described with line references after my general comments.

The main issue concern the originality of this study. This research is not a novel idea in SO₂ measurements at active volcanoes by scanning spectrometer system. In particular, the pioneering application of a modelled FRS in NOVAC data at the Piton de la Fournaise has been not mentioned and/or discussed (Hibert et al., 2015). In this work, the authors successfully retrieved SO₂ CD and flux from the NOVAC scanning network without invoke different ILS as in the manner of Salerno et al., 2009a and no laborious PCA analysis for accounting of instrumental effects in the spectrometers. Nevertheless, Lubcke et al., set the originality of their study in the PCA analysis. The authors carried out this statistical test in free-volcanic gas spectra selected from the database of Nevado del Ruiz and Tunguraha NOVAC network. Selected 7-days data between Sept-Oct at Nevado del Ruiz and 10-days/year between 2009 and 2011 at Tunguraha. Results, were assumed reflecting the instrumental features of the entire network. However, as reported by Pinardi et al., 2007, instrumental effect dramatically

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may change over time due to temperature drift, and these are different between spectrometers (eg., Instrumental line function, stray lights). As the NOVAC instruments are not thermo-stabilised (Galle et al., 2010) variable instrumental drifts may take place at different magnitude and time scale. This raises the hesitation to believe that the PCA results gathered from few data and from a unique spectrometer (per network) may be assumed representative of the instrumental features of entire scanning networks. The authors, attempt to explore this issue by comparing the residual structures and optical depth (Fig. 3, 4, and 5) obtained by retrieving the entire records of data from the two networks. They found a reasonable agreement between the instruments. This find arise anyway a further question. In detail, since the results of the comparison relates to the PCA results and as instrumental drifts are taking place physically in different way, the question that come up is whether the statistical PCA has a fundamental role in the framework of the paper.

There are also some comments about the structures of the article. The paper needs to be restructured, reducing the number of paragraphs and length of the article itself. For instance, the detailed description of the DOAS technique is unnecessary. This technique is a very well established spectroscopy approach and the theory largely published. I really suggest the authors to replace details and equation with references (Platt and Stutz, 2008). Moreover, some paragraph might be eliminated or merged with others, e.g 4.2 consist of only five lines. Instead, I'd suggest the authors to include a small paragraph describing the NOVAC networks object of the study. Finally, a graph of the SO₂ CDs retrieved by FRS throughout the years at both Nevado del ruiz and Tungurahua should be displayed for completeness of the study.

Other comments Abstract: Pag 1, Section 05, line 1: define here the scanning DOAS, change this sentence with '.....scanning ultraviolet spectrometers network, also named as scanning DOAS '. Pag 1, Section 10, line 3: specify volcanoes. Pag 1, section 20, line 1: 'complicated instrumental calibrations' please report which kind of calibrations are required in field 'eg.,.....' . Pag 2, section 05, line 3-4: It's not clear if

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the word 'New', is related to the FRS or the entire study carried out in the paper. In case it relates to the FRS method, as mentioned in the general comment, I'd suggest to remove 'new' because it is not a novel approach.

Introduction Pag 2, section 10, line 2: add reference Williams-Jones, et al, 2008 and Oppenheimer, 2011 Pag 2, section 10, line 2-3: More recently the availability of miniature spectrometers allowed.': I suggest to delete more recently because it's now more than twelve years after the first application of this technique at active volcanoes by Galle et al., 2003. Pag 2, section 10, line 5: add reference 'Elias et al, 2006. Pag 2, section 15, line 2-3: change 'One of the first installations were scanning DOAS instruments at Montserrat volcano (Edmonds et al., 2003)' with 'The first installations of scanning-DOAS network was developed at Montserrat volcano (Edmonds et al., 2003)'. Pag 2, section 20, line 1: add reference Salerno et al., 2009b. Pag 3, section 15, line 1-4: specify large NOVAC database of the two Colombian and Ecuadorian volcanoes object of this research. Pag 3, section 30, line 5-7: move and change 'First steps towards the here described approach were taken in Lübcke (2014), where measurements from NOVAC instruments at Nevado del Ruiz were evaluated for SO₂ with a modelled background spectrum'. With 'this approach was successively adopted by Lübcke (2014) and Hibert et al., 2015 for evaluating NOVAC data collected at Nevado del Ruiz and at Piton de la Fournaise Reunion, respectively'. Pag 3, section 10-15, line 3-3: move and rephrase this section in Pag. 2 section 25-30 while talking about the use of the modelled FRS. Report that limitations and uncertainty of the standard NOVAC retrieval has been already discussed and overtaken applying a modelled background FRS by Hibert, C., et al. 2015.

2 Background spectra for scanning DOAS instrument networks at volcanoes

As reported in the general comments, the theory behind the DOAS evaluation is widely published. Thus allocate paragraph to a detailed description of the physics/equation of DOAS does not provide any advantages to the article. I'd suggest to replace part of this section with Platt and Stutz, 2008. Pag 4, section 30, line 7-8: The experiment

reported in Salerno et al., 2009a does not imply any routinely specific spectroscopy retrieval operations to be performed in field. SO₂ calibrated quartz cells spectra were employed for exploring and validate at different timescale the application of a modelled FRS for reducing ultraviolet open-path spectra from scanning ultraviolet spectrometers. The comparison of three year of SO₂ CDs an flux form scanning system and traverses (Salerno et al., 2009b) provide a further quantitative constrain on the efficiency of the approach developed in Salerno et al., 2009a. Pag 5, section 10, line 1-5: rephrase this section and delete math equations. Pag 5, section 15, line 4-5: As reported in the paper (pag 6, section 20) SO₂ retrieval was performed between 310.0 - 326.8 nm, report the reason for which high resolution laboratory cross-sections were convolved using the Hg line at 334.15 nm and not for instance 313.16 nm.

3 Data evaluation 3.1 Settings of the DOAS retrieval

Pag 6, section 15, line 5: 'for both retrieval methods (see below)' define here the two retrievals. Pag 6, section 25-30, line 4-9: detail of standard NOVAC retrieval has been already reported in the introduction, delete this section or synthesise. Pag 7-8, section 30 – 30: replace DOAS theory with references.

3.3 Principal Component Analysis for Method B Pag 9, section 10-20, the authors report that PCA needs to be run using clean-gas spectra. However, due to volcanic activity this is not so straightforward. Therefore, only few dataset of spectra were selected from both Nevado del Ruiz and Tungurahua database. Nevertheless, at Pag 14 paragraph 4.3 the requirements of clean-gas spectra arise again for evaluating the sensitivity of the retrieval as zero CDs. As reported in this paragraph at section 5, line 2-4 a greater number of data-days were discriminated at both volcanoes respect what have been selected for the PCA analysis (e.g, at Nevado del Ruiz 7 days vs 73 days). Please, rephrase the two section in case of mistake or clarify this conflict issue. Pag 9, section 15, line 5: constrain in a quantitative scale the meaning of 'little degassing activity'. Pag 9-10, section 30-5, define period of selected data at Tungurahua. Pag 10, section 30, line 7: constrain in a quantitative scale the meaning of 'low volcanic degassing activity'.

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3.4 Implementation of the new algorithm

I would suggest the authors to remove this paragraph. It is a summary of what has been already reported in the manuscript.

4 Results

Pag 11, section 25, line 1: delete brackets in Lubcke et al., 2013. Pag 14, section 05: see comments Pag 9, section 10-20. Conflict or erroneous explanation of this issue. Pag 17-18, section 05. Method A and B retrieve the same spectra, explain in more detail why the effect of stray light should affect the two retrieval performed with method A and B in a different way.

4.2 DOAS fit example This paragraph consist of only five lines, please remove it or include in a different paragraph.

Figure Fig 9: change 'which is similar to the standard NOVAC approach' with 'standard NOVAC approach'.

Litterature Elias, T., Sutton, A.J., Oppenheimer, C., Horton, K.A., Garbeil, H., Tsanev, V., McGonigle, A. J.S., Williams-Jones, G., 2006. Intercomparison of COSPEC and two miniature ultraviolet spectrometer systems for SO₂ measurements using scattered sunlight. Bull. Volcanol. 68, 313–322.

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Lübcke, P., et al., 2015, DOAS evaluation of volcanic SO₂ using a modeled background spectrum: Examples from the NOVAC stations at Nevado del Ruiz (Colombia) and Tungurahua (Ecuador), *Geophysical Research Abstracts* Vol. 17, EGU2015-1803-1, EGU General Assembly 2015, Vienna.

Hibert, C., A. Mangeney, M. Polacci, A. D. Muro, S. Vergnolle, V. Ferrazzini, A. Peltier, B. Taisne, M. Burton, T. Dewez, G. Grandjean, A. Dupont, T. Staudacher, F. Brenquier, P. Kowalski, P. Boissier, P. Catherine, and F. Lauret (2015), Toward continuous quantification of lava extrusion rate: Results from the multidisciplinary analysis of the 2 January 2010 eruption of Piton de la Fournaise volcano, La Réunion. *J. Geophys. Res. Solid Earth*, 120, 3026–3047. doi: 10.1002/2014JB011769.

Oppenheimer, 2011, Ultraviolet sensing of volcanic sulfur emissions, *Elements*, Vol. 6, pp. 87–92, DOI: 10.2113/gselements.6.2.87.

Salerno, G. G., M. R. Burton, C. Oppenheimer, T. Caltabiano, D. Randazzo, N. Bruno, and V. Longo (2009b), Three-years of SO₂ flux measurements of Mt. Etna using an automated UV scanner array: Comparison with conventional traverses and uncertainties in flux retrieval, *J. Volcanol. Geotherm. Res.*, 183, 76–83, doi:10.1016/j.jvolgeores.2009.02.013.

Williams-Jones, G., Stix, J., Hickson, C., 2008. The COSPEC Cookbook: making SO₂ measurements at active volcanoes. IAVCEI, *Methods in Volcanology*, vol. 1. and Oppenheimer, 2011, Ultraviolet sensing of volcanic sulfur emissions, *Elements*, Vol. 6, pp. 87–92, DOI: 10.2113/gselements.6.2.87.

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