Atmos. Meas. Tech. Discuss.,
doi:10.5194/amt-2016-24-RC4, 2016

Atmospheric Measurement Techniques Discussions

Interactive comment on "Retrieval of absolute SO_2 column amounts from scattered-light spectra – Implications for the evaluation of data from automated DOAS Networks" *by* Peter Lübcke et al.

Anonymous Referee #4

Received and published: 6 June 2016

This paper presents an approach to analyse UV scattered sunlight spectra of volcanic plumes, such that an absolute rather than relative quantification of SO2 slant column amounts can be achieved. The motivation for this approach is that the NOVAC network of scanning UV spectrometers has used since its inception an approach where a reference spectrum from within each scan is used to remove solar spectrum features. SO2 values are then offset by the minimum SO2 value in each scan. This has the advantage of excellent solar feature removal, but also means that if SO2 is ubiquitous then the minimum SO2 value will not represent zero SO2, and an underestimate of column amounts will affect the scan, leading to flux underestimates. On the contrary, by using an absolute method for SO2 retrieval this problem is addressed.

The approach proposed is effectively identical to that reported in Salerno et al. 2009a, which has been applied successfully by INGV Italy to the scanning UV spectrometer SO2 flux monitoring networks on Etna and Stromboli since 2006. Salerno 2009a used a high resolution UV solar spectrum (Kurucz et al., 1984) to model the Fraunhofer features of the solar spectrum, whereas Lubcke et al. use the Chance and Kurucz (2010) Solar atlas.

Salerno et al., 2009b examined three years' results using the algorithm described in Salerno et al., 2009a from the automated flux monitoring network on Mt. Etna with traverse flux measurements, which are in many ways much more robust than scanning flux measurements: a clear sky spectrum can be collected which is guaranteed to be SO2 free, and there are no geometric corrections to make for plume height.

Burton and Sawyer 2016 present an update to the Salerno et al. 2009a work in which they recognise that there are instrumental response features which add a fixed pattern to the measured spectra using CCD-based spectrometers, the 'flat' spectrum. This adds 1-3% noise but can be readily characterised and removed from each spectrometer using lab-based measurements of a broad-band UV source.

The issues associated with the standard NOVAC analysis system have been well known for over a decade, since the implementation of the approach of Salerno et al. 2009a. The main objective of the Lubcke paper is that (page 3 line 1-3) "This work will follow the idea of using a high resolution Solar atlas spectrum (Chance and Kurucz, 2010) in order to calculate a gas free background spectrum which is used as an FRS for the DOAS evaluation of SO2.". It appears however that this work has already been done by Salerno et al. 2009, so the novelty of the current work should lie elsewhere. This immediately highlights that a major refocussing of the manuscript is needed, because so much of the work has already been developed.

A valuable contribution of the Lubcke paper is the comparison of the SO2 offset correction with the Salerno et al. 2009a type retrieval for data from Nevada del Ruiz and

Tungurahua. However, with 30 volcanoes in the NOVAC network and the fact that an established method for dealing with the issues of absolute SO2 retrievals has been applied for over a decade, it seems wholly inadequate that only two volcanoes are investigated. My recommendation is therefore to refocus this work from a rehash of previously published methods to a thorough investigation of the full implications of the application of the SO2 offset approach to the NOVAC network. Sincerely, with the time available since this issue was highlighted and a solution shown it is not acceptable to present just for two volcanoes from 30. A further requirement for a revised or re-submitted paper is that some comparison with traverse flux data is used. The final objective of all this work is to get as accurate as possible SO2 flux data, not SO2 slant column amounts. The corrections posed may have an angular dependence, leading to unexpected impacts on the reconstructed SO2 fluxes from scans. The only way to evaluate this properly is to compare scanner SO2 fluxes with traverse flux measurements, as performed by Salerno et al., 2009b, which should also be referenced here. I would therefore strongly recommend that such measurements and their comparison be included in a future revision, as otherwise the veracity of the final SO2 flux measurements will remain doubtful.

The final contribution of the Lubcke paper is a PCA analysis which effectively does a qualitative job of removal of the flat spectrum as described by Burton & Sawyer 2016. The PCA approach means the user absolves themselves of responsibility for finding the physical process producing the observed features, which is rather disappointing, as precise physical understanding of the measurement process is how we can improve in the future. The majority of the features captured by PCA are flat spectrum, so this contribution is somewhat redundant, given that a clear physical explanation of the process is given by Burton & Sawyer 2016.

The present work is also overly long with a redundant exhaustive explanation of the basic retrieval technique. Significant shortening is essential.

Burton, M. R. and Sawyer, G. M.: iFit: An intensity-based retrieval for SO2 and

BrO from scattered sunlight ultraviolet volcanic plume absorption spectra, Atmospheric Measurement Techniques Discussions, 2016, 1–47, doi:10.5194/amt-2015-380, http://www.atmos-meas-tech-discuss.net/amt-2015-380/, 2016.

Kurucz, R.L., Furenlid, I., Brault, J.,and Testerman, L., 1984. Solar flux atlas from to 1300 nm, Natl. Sol. Obs., Sunspot, New Mexico, 240.

Salerno, G.G., Burton, M., Oppenheimer, C., Caltabiano, T., Tsanev, V.I., Bruno, N., 2009a, Novel retrieval of volcanic SO2 abundance from ultraviolet spectra. J. Volcanol. Geotherm. Res. 181, 141–153. doi:10.1016/j.jvolgeores.2009.01.009.

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

Interactive comment on Atmos. Meas. Tech. Discuss., doi:10.5194/amt-2016-24, 2016.