**Review of J. Gliß et al., OClO and BrO observations in the volcanic plume of Mt. Etna – implications on the chemistry of chlorine and bromine species in volcanic plumes**

**General Comments and Recommendation**

♦ The description of the formation of BrO in volcanic plumes is not full – e.g. the reaction involving ultraviolet radiation is missing. This reaction is the basis for the explanation of the increase of BrO early in the morning . A very good description of formation of RHS in volcanic plumes is presented by L. Sulf et al. (this journal 14, 23639-23680). At the same time the formation of BrO is regularly explained in all papers reporting BrO in volcanic plumes and in the original papers of Wennberg, von Glasow and others. Why not just provide a reference?

♦ The estimation of BrO and OClO concentration is based on the assumption of circular plume but all cross sections reported in the manuscript (c.f. figures 7 and 8) are strongly suggesting that plume has a form quite different from the circular. Thus the reported values are rather not realistic.

♦ The statement that it is possible to measure and to estimate corresponding mean and standard deviation of some parameter below the detection limit is nonsense from statistical point of view (c.f. figures 7, 8, and 10). This means that the threshold definition is wrong or threshold limit itself is not estimated properly.

♦ The reported increase of $X_mY_n/SO_2$ ($BrO/SO_2$ and $OClO/SO_2$) ratios at the edges of volcanic plumes is a tendency that needs further verification at different atmospheric conditions and in plumes of different volcanoes. The reported results are not totally convincing. Plume edges in figure 8 look rather as tails of the plume caused by its drifting due to the wind variations. The quite long time required for collection of a single spectrum (2.5 minutes) and the scanning angle increase by 4 degrees are also prompting for probable missed plume structure. Also, in cases

when it was possible to access plume edge by scans perpendicular to plume direction, an increase of $BrO/SO_2$ is observed only in 30±18% of the cases whilst decrease or no change in 16% and 8% respectively. This means that it is possible to talk about tendency only and further investigations are necessary.

♦ The reported stratospheric BrO column amounts are up to two times larger that the mean values reported previously by other authors. Is it possible that this fact is compromising the reported variation of BrO early before sun rise?

♦ The description of DOAS retrieval will arise many question by DOAS users. I suggest that part of the manuscript should be significantly improved by paying particular attention to the following:

(1) It is stated that the used spectrographs is Avantes 5 AvaBench-75-Ultra Low Straylight. Probably the authors refer to AvaBench-75-ULS-2048. This means there are about 6 pixels per instrumental slit function in UV (0.51 nm) and VIS (0.31 nm). These numbers are on the limit when it is necessary to account for undersampling effect. Please elaborate its ignoring.

(2) Please explain how the absorption cross sections have been corrected for saturation effect.

(3) Why it is necessary to perform $I_o$-correction when convolving all absorption cross sections (especially the strongest $SO_2$ one) included in the fit. Originally the $I_o$-correction was used to correct the fit of weak absorbers (stratospheric BrO).

(4) The origin of R4 Ring effect and the improvements achieved by its usage.

(5) Explain necessity of using two $O_4$ absorption cross-sections (table 2).

(6) The used absorption cross sections are measured by different authors and are loaded with individual errors (especially with respect to wavelength calibration). As a result linking the shift of all cross sections included in any particular fit to the shift of the strongest one could be a quite arguable

decision. Please elaborate the reasons.

(7) Explain the meaning of zero-order polynomial and the improvements achieved by its usage. Is this actually the offset polynomial?

(8) Why formaldehyde is included in the fit. Explain the origin of $H_2CO$ in volcanic plumes.

(9) The three gases of interest $SO_2$, BrO and OClO are fitted in three different fitting windows but these gases are abundant in the same plume at the same time instance, i.e. the retrieved $SO_2$ column amount has to be the same in all fitting windows. To my knowledge only WinDOAS and Q-DOAS are capable to perform the fit in this manner. How this issue was resolved when using DOASIS.

(10) The radiative transfer effects (RTE) are totally ignored in this work. According to figure 1 some measurement locations were more than two kilometres from the Etna's craters. The distances to the plume have to be reported for each location and the neglecting of RTE has to be explained in details. Please refer to Kern et al., 2008, who proved the necessity to account for RTE in all DOAS retrievals. The fact that RTE effects are ignored in this manuscript may be accepted in a very wrong way – recommendation just to ignore them as unnecessary complication when interpreting the retrieval results.

**Specific Comments**

(1) Provide reference for MS-DOAS software or describe it briefly.

(2) Explain how a UV camera may be used to estimate the wind direction – UV camera is registering 2D-projections.