Reply to Reviewer #1

We are thankful to Reviewer #1 for the constructive comments. His introductory remarks/questions have been grouped as 3 general questions. To these 3 general questions, as well as to his additional 17 specific comments, our responses and revisions in the text are as follows:

General question 1: Reviewer #1 criticizes our overlooking of the difficulties to see particularly in the OMI data the volcanic SO_2 signals seen by the Brewers and as he points out "The authors need to temper their conclusions ….".

Response to general question 1: Reviewer #1 correctly points out that we should have tempered our conclusions concerning the SO_2 excursions following large volcanic eruptions because they could not be seen equally well in the OMI and GOME-2 satellite measurements as was the case with the Brewer network, except for Kasatochi. We have carefully revisited the OMI and GOME-2 data sets and found out that during the most perturbed period following the eruptions of Bardarbunga and Eyjafallajökull the satellite measurements from overpasses were so sparse that the daily average was not corresponding to the Brewer network sample. For instance and following Bardarbunga and Eyjafjallajökull, there were many days where we had only one or two OMI overpassing measurements following the eruption, obviously not representing the 19 Brewer instruments in Europe. To temper our past conclusions we have applied a criterion (see new section 3.1) according to which "a daily average from either OMI or GOME-2 should be calculated if and only if more than half of the individual overpasses had data at a given day". As can be seen from the revised figures 4 and 12, OMI data are missing for not meeting this criterion. The only firm conclusion that can be drawn with statistical confidence is that from all three eruptions with volcanic SO_2 plumes overpassing the Brewer network and seen as well from OMI and GOME-2, a strong positive signal can be confirmed only in the case of Kasatochi eruption (we have redrawn the time series, see new Fig. 13). Following these major changes, we have rephrased our abstract and conclusions accordingly.

General question 2: "The most serious issue is why there is such poor correlation of the satellite data, particularly OMI, with the Brewer data for 4 out of the 5 eruptions compared, and then such good agreement with Kasatochi? Was there something different about Kasatochi? When there is such poor agreement I don't see the point of quoting averages of the satellite data which appear to this reader to be in the noise of the measurements".

Answer to general question 2: Indeed as mentioned above the best agreement was found for the case of Kasatochi because it happened to have many measurements from coinciding satellite overpasses during common days with the Brewer instruments. For the case of Bardarbunga and for the case of Eyjafjallajökull, the satellite data were sparse, particularly for OMI. For Bardarbunga, the correlation between the GOME-2 overpasses and Brewer stations under the volcanic SO_2 plume was calculated to be 0.44, statistically significant at the 99% confidence level in spite the fact that during the two days of peak SO_2 levels (21-22/9/2014) as "seen" at the Brewer stations, there were no satellite data available. For Eyjafjallajökull similar sparsity of the data reduces confidence and unfortunately for OMI we could not calculate correlations with the Brewers at all due to the small sample of the satellite data. We note here that the case for Grimsvotn volcano has been removed as recommended by reviewer #2 comments and is not discussed in the revised paper. The reason is that the volcanic SO_2 plume has been always outside of the Brewer network. The text has been revised in concurrence to the above findings.

General question 3: The reviewer points out the problem in measuring SO_2 columns, where to set the zero point as well as what is the meaning of negative SO_2 columns and how to interpret them and related questions on the noise, the baseline and the correlations in figures 5, 10, 14, 15 and 16.

Answer to general question 3: In the text (section 2.1) we have added a full description of the Brewer algorithm and the reasoning on the existence of some negative values which could be considered either as small or as noise. The text now reads: "From the above described operational Brewer algorithm it is evident that the estimation of columnar SO_2 is the result of the difference between two columnar terms (O_3 + SO_2) and O_3. Both terms have uncertainties (weighting functions, calibrations, random errors, systematic errors). Systematic negative values could be the result of a systematic offset in the measurements that can be related to the calibration of the instrument (usually optimized only for the ozone measurements). Randomly varying positive and negative values around zero, suggest that the signal of SO_2 is small (and thus the difference of two terms should be close to zero) but since both terms have uncertainties, negative values are possible indicating that the amount of SO_2 in the atmosphere is below the detection limit of the instrument and could be considered as noise. In this work we have repeated our analysis excluding the negative values and the results remained the same i.e. a positive increase after a major volcanic eruption was confirmed as described in the following sections".

After careful consideration, we decided to recalculate all values and redraw all Brewer composite figures by considering that 10 days before the volcanic eruption all Brewer and satellite observations obviously did not contain any volcanic signal. The data set which included daily values during the 10-day unperturbed period before the eruption, was considered to represent the base line for each Figure. Subsequent grouping in the **new Figures** (4, 9, 12, 13) show the departures of mean SO_2 columns from the unperturbed baseline and all numbers in Table 4 have been recalculated as departures from the unperturbed 10-day baseline.

Answers to specific comments

Comment 1: "1.41-42. Have increased compared to what? That so2 columns increase following somewhat large volcanic eruptions is not new and has not depended on this paper to show that. Nor is it new that such columns increased following the five eruptions considered here. This sentence needs to be rephrased or deleted. I would begin the abstract with something like.
Following the five largest volcanic eruptions of the past decade in the Northern Hemisphere, a strong positive SO_2 signal was detected by all the existing networks either ground based (Brewer, EARLINET, AirBase) or from satellites (OMI, GOME-2). This study particularly examines …
But after reading the paper even this sentence has issues. A strong signal was not detected in OMI and GOME-2 data according to the results shown here in several cases. Thus the statement that a "… a strong positive SO_2 signal was detected by all the existing networks either ground based (Brewer, EARLINET, AirBase) or from satellites (OMI, GOME-2) …" is not correct for the satellite data for all cases".

Answer to comment 1: In the revised text we clarify that the SO_2 columns have increased relative to the unperturbed 10-day baseline. We also specify that a strong positive signal was detected by all the existing networks only at Kasatochi. As mentioned before, the abstract and conclusions have been fully revised accordingly.

Comment 2: "1.41. Why are the increases described as significant? Significant in what way? The so2 increases following Pinatubo and El Chichon were significant, but these are on a different scale than the eruptions considered here".

Answer to comment 2: In the revised text the increases are described as departures from the ten days before the eruption where all Brewer and satellite SO_2 measurements are considered as non-perturbed. A departure was characterised significant if it exceeded 3σ, where σ was calculated from all daily values 10 days before all eruptions and for as many locations as the number of the measuring stations or the corresponding satellite overpasses in the cases of OMI and GOME-2.

Comment 3: "1.45-47. This statement is incorrect for the reasons given above, particularly for OMI. The correlation is better for Brewer and GOME-2, but I doubt even this would be statistically significant at the level claimed if all cases were considered. See Figs. 5, 12, 15. Again how are the columnar so2 amounts significant? What do the authors intend to imply with this word?"

Answer to comment 3: In our original manuscript sparsity of data from OMI and to a lesser extent from GOME-2 resulted to wrong correlations with the data from the Brewers. In the revised text the correlations between the Brewers and GOME-2 have been corrected and were estimated to be 0.31 (95% confidence level) and 0.44 (99% confidence level) in Eyjafjallajökull and Bárðarbunga, respectively. Correlations between the Brewers and OMI were not calculated due to the scarcity of OMI data in Eyjafjallajökull and Bárðarbunga (see corrected Table 5, corrected text and abstract).

Comment 4: "3.9-14. The authors need to be more careful about their claims concerning the "five" volcanic eruptions. In the abstract it was the 5 most significant eruptions since 2005. Now here it seems to be the five eruptions which produce the most so2 over Iceland, but only 4 eruptions are shown. Not surprisingly 3 of these eruptions were in Iceland, although most of these eruptions are not on the list of the 5 eruptions since 2005 with the greatest atmospheric impact. Here the sentence needs to indicate up front that these are selected based on their so2 columns over Iceland. So … Five cases of high SO_2 over Iceland from volcanic …
Yet this sentence goes on to say that these are the five eruptions to be compared in this study. So I am confused, are the eruptions the 5 most significant since 2005 or the 5 with most significant so2 over Iceland. According to the Smithsonian Global Volcanism Network, Bárdarbunga has a VEI of zero, so undetermined.
Table 1. It is significant that 4 of the 5 eruptions are at high northern latitudes, while the lone tropical eruption had its plume picked up in the Asian monsoonal circulation to bring the so2 over Europe, so an important but poorly stated criteria seems to be the opportunity to measure the plume over Europe.

Clearly there is enough confusion here that the authors need to rethink the criteria used for the selection of the 5 eruptions and to explain it clearly".

Answer to comment 4: We consider all major eruptions that have occurred in the N.H. in the past decade according to the Smithsonian Global Volcanism. The text has been revised and reads now as follows:

"Table 1 lists in chronological order all major volcanic eruptions in the Northern Hemisphere between 2005-2015 with volcanic explosivity scale index (VEI) of at least 4 (Newhall and Self, 1982; Robock et al., 2000; Zerefos et al., 2014). The study also provides a separate analysis for the Bárðarbunga eruption, which although not rated 4 has been already studied with the Brewer at Sodankylä by Ialongo et al. (2015).

As seen from Table 1, chronologically, the first case was the volcanic eruption at Mount Okmok, Alaska (53.43^o N, 168.13^o W, 1073 m above sea level (asl), 12 July 2008, Prata et al., 2010) followed by the Kasatochi eruption, Alaska (52.17^o N, 175.51^o W, 300 m asl, 7-8 August 2008, e.g., Kristiansen et al., 2010; Krotkov et al., 2010; Waythomas et al., 2010) which was detected over large areas of the Northern Hemisphere. Okmok and Kasatochi volcanoes in Alaska erupted a short time span of less than a month and therefore we decided to study the evolution of the Brewer SO_2 columnar measurements following the latter volcanic eruption (Kasatochi). The third eruption took place at Sarychev in Russia (48.1^o N, 153.2^o E, 1496 m asl, 12-17 June 2009, Haywood et al., 2010). The evolution of the SO_2 volcanic plume from Sarychev was mostly observed over the North Pacific, North America and North Atlantic (Haywood et al., 2010). There was only one North American Brewer station (Saturna Island) in the path of the plume from Sarychev eruption. The record shows SO_2 columns of 8.6 DU detected on 19 June 2009 and 3.7 DU on 20 June 2009. This volcanic eruption is not investigated any further in this paper. The next eruption on the list, Eyjafjallajökull in Iceland (63.63^o N, 19.62^o W, 1666 m asl, from 14 April to 23 May 2010), resulted in interruption of the air traffic over NW Europe (e.g. Flemming and Inness, 2013). The fifth eruption Grímsvötn 2011 (64.42^o N, 17.33^o W, 1725 m asl, 21 May 2011) was studied by Flemming and Inness (2013), and by Moxnes et al. (2014). This eruption provided an interesting example of a clear separation of the volcanic SO_2 plume (transported mostly northwestward) while the fine ash was transported mostly southeastward. Unfortunately the volcanic plume did not overpass any Brewer station and therefore we do not include any results post Grímsvötn eruption. The sixth eruption recorded features the Nabro in Africa (13.37^o N, 41.70^o E, 2218 m asl) that occurred on 12-13 June 2011 (e.g., Bourassa et al., 2012; Sawamura et al., 2012; Clarisse et al., 2014). We present here a case study that described detection of the Nabro volcanic SO_2 plume over ground based stations. The plume was clearly detected by the Brewer instrument over Izaña (and poorly from space), then over Taiwan by both Brewer and satellite instruments, and finally at Mauna Loa, Hawaii (mostly by the Brewer instrument). The seventh eruption was Tolbachik, Russia (55.83^o N, 160.33^o E, 3.611 m asl) on 27 November 2012 (e.g. Telling et al., 2015). As in the case of Grímsvötn, the plume has not passed over any Brewer station that was verified by trajectory analysis. The next eruption on the list is the volcanic eruption from Bárðarbunga, Iceland (64.64^o N, 17.56^o W, 2005 m asl) that was observed between 31 August 2014 and 28 February 2015 (e.g. Schmidt et al., 2015). This last eruption, although not yet rated on the VEI scale, has been extensively studied in view of the observed increased SO_2 concentrations that have been observed all the way through troposphere and reaching down to the surface in Europe (Ialongo et al., 2015; Schmidt et al., 2015)."

Comment 5: "4.13-21. Confusing. I had to re-read this several times. First the authors state … the Brewer spectrophotometer is additionally used to derive the SO_2 column.., Then they

say … The existing Brewer network could deliver frequent SO_2 measurements as well, but the Brewer instruments are less able to accurately provide SO_2 measurements … So which is it? Don't claim that it is used and then say it can't be used. Please rewrite this to be clear".

Answer to comment 5: The sentence has been rewritten and reads as follows: "Because sulfur dioxide has strong and variable absorption in this spectral region, the Brewer spectrophotometer has additionally been proposed to derive SO_2 columns (Kerr et al., 1980). About two hundred Brewer spectrophotometers around the world contribute high-precision ozone data to the global ozone monitoring network (Kumharn et al., 2012). The existing Brewer network also delivers frequent SO_2 columnar measurements as well, which can be used for analyses, but with caution". (See revised section 2.1).

Comment 6: "7.2-4. Doesn't this also suggest a bias for the Brewer data?"

Answer to comment 6: Any biases in the data have been eliminated by expressing all data (Brewer, GOME-2 and OMI) as departures from the unperturbed 10 day period prior to the volcanic eruptions. The new text now reads: "Averaging the departures from the pre-volcanic baseline for all Brewer stations and for all bimonthly periods gives a mean SO_2 columnar departure of 0.10 ± 0.03 DU. This estimate is on the same order of magnitude as the corresponding statistics for OMI (TRM) SO_2 column departures (0.05 ± 0.02 DU) and that measured by GOME-2 (0.09 ± 0.02 DU)".

Comment 7: "7.35-36. From Fig. 5 only the GOME-2 measurements corroborate the Brewer results, but even then only in timing, not in magnitude. Is there an explanation why no signal appears in OMI data and why the Brewer and GOME disagree in magnitude to the extent shown?"

Answer to comment 7: The explanation is the sparsity of OMI and GOME-2 data, particularly OMI, during the days of elevated SO_2 column observed by the Brewer network. Figure 5 (new figure 4) has been redrawn by applying a criterion according to which a daily average from either OMI or GOME-2 should be calculated if and only if more than half of the individual overpasses had data at a given day. The text has been revised and reads now as follows: "As shown in Figure 4a, the SO2 plume was detected by the Brewer instruments located in the passage of the volcanic SO2 plume and from different ground based networks. However, no co-incident measurements were available from the OMI and GOME-2 overpasses at the time of the high SO2 excursions".

Comment 8: "8.23-30. Aside from GOME-2 it seems pointless to quote these numbers for OMI. The OMI data do not indicate anything out of the ordinary for 20-25 September, neither the TRM nor PBL. In fact there are bigger excursions of the so2 column at other times. The GOME-2 data are better and a case can be made that some so2 was observed, but even these data could be questioned".

Answer to comment 8: In the revised text we do not quote these numbers for OMI. The new text now reads: "As can be seen from Figure 4a, the highest SO_2 column departures from the pre-volcanic baseline were observed from 21 to 22 September 2014. The mean SO_2 column measured by the Brewers under the plume was 2.4 ± 0.8 DU, which was five times greater than the mean column of SO_2 measured by the Brewers outside of the plume (-0.1 ± 0.1 DU) by 2.5 DU on average. The "error bars" show the standard deviation of the daily SO_2 values of all stations during the non-perturbed 10 day period prior to the volcanic eruption. These differences provide rough estimates of the additional SO_2 loading induced by the volcanic eruption over Europe which exceeds 3σ. Comparison between satellite data and Brewer are limited for interpretation because satellite measurements are sparse, represent an average SO_2 column over a relatively large satellite pixel, while the Brewer observations are designed to provide a local point measurement".

Comment 9: "8.33-35. Thus the statement, "In all cases, however, the observed … were always higher …" is simply incorrect, as demonstrated with the numbers just above, and should be removed".

Answer to comment 9: The statement has been removed.

Comment 10: "9.1-5. Why is there so much inconsistency between Figures 5 and 7. Fig. 7 shows OMI measurements of 1-4 DU across large regions of Europe, yet Fig. 5 indicates almost all OMI measurements < 1 DU and most measurements < 0.5 DU".

Answer to comment 10: We would like to clarify that Figure 7 (now has become Fig. 6) does not show OMI measurements but forecasted calculations by the MACC model with and without OMI assimilation for 21 September 2014. On the other hand Fig. 5 (now has become Fig. 4) is based on actual measurements, in which OMI had only a couple of measurements over the Brewer sites.

Comment 11: "Figure 9. The differences between the coloured lines are not obvious".

Answer to comment 11: The figure has been redrawn to become clear.

Comment 12: "10.15. What is meant by both methods?"

Answer to comment 12: "It is clearly shown that the zero-calibrated Brewer SO_2 data do not compare well with OMI and GOME-2 levels. Instead, the Langley calibrated Brewer data compare better with OMI and GOME-2 retrievals". This is clarified in the new text (see section 3.2, page 12, new lines 22-24).

Comment 13: "Fig. 15. Why is the Brewer baseline at 0.2-0.3 DU for the stations under the plume, whereas for the 10 outside stations the baseline is closer to zero?"

Answer to comment 13: It has to do with the offset of the instruments. We have overcome this problem by analysing departures from the non-perturbed ten days prior to the eruption as described before. The new Figure 12 (old figure 15) does not show this discrepancy anymore.

Comment 14: "11.38. Does an average SO_2 plume of 0.1 DU mean anything when earlier the averages of the Brewers without influence by volcanoes was on the order of 0.4 DU? It does not help the authors' argument to be calling out numbers in the text which are in the noise of the measurements. The authors also never explain what a negative DU measurement means. What causes this? Are the negative numbers a real measurement?"

Answer to comment 14: No, it does not mean anything. All SO_2 columns have been recalculated as departures from the non-perturbed 10-day baseline and we do not call out numbers which are in the noise of the measurements as can be seen in the new text (section 3.3).
With regard to the negative SO_2 columns, we clarify in the revised section 2.1 that "From the above described operational Brewer algorithm it is evident that the estimation of columnar SO2 is the result of the difference between two columnar terms (O3 + SO2) and O3. Both terms have uncertainties (weighting functions, calibrations, random errors, systematic errors). Systematic negative values could be the result of a systematic offset in the measurements that can be related to the calibration of the instrument (usually optimized only for the ozone measurements). Randomly varying positive and negative values around zero, suggest that the signal of SO2 is small (and thus the difference of two terms should be close to zero) but since both terms have uncertainties, negative values are possible indicating that the amount of SO2 in the atmosphere is below the detection limit of the instrument and could be considered as noise. In this work we have repeated our analysis excluding the negative values and the results remained the same i.e. a positive increase after a major volcanic eruption was confirmed as described in the following sections".

Comment 15: "Fig. 16. Why is a 7 day running mean now added to the measurements? Does it show something missing in the simple averaged daily data shown up to now?"

Answer to comment 15: To avoid confusion the left panel of that figure has been removed. Please note that the new figure for Kasatochi is now Fig. 13 because the paragraph for Grimsvötn has been removed as requested by Reviewer #2.

Comment 16: "12.31-13.6. A calculation of Pearson's correlation coefficients is not necessary to convince the readers that the Brewers, GOME-2 and OMI are all in agreement at least over Europe. Is the Taiwan station included in the correlation coefficients? If so, does the fact that there is virtually no correlation there get masked because it is only one station? What is telling about this paragraph, and the corresponding Table 5, is that such tests were not used in any previous comparison, most certainly because the results would have been much worse, see Figures 5, 12, 15".

Answer to comment 16: No, Taiwan is not included in the correlation coefficients. Table 5 has been redrawn to show the correlation coefficients between the Brewers, GOME-2 and OMI over Europe in all three volcanic eruptions (Kasatochi, Eyjafjallajökull and Bárðarbunga). The correlations between the Brewers and GOME-2 were found to be statistically significant in all volcanic eruptions. Brewer and OMI data were strongly correlated in Kasatochi but unfortunately the sparsity of OMI data during Eyjafjallajökull and Bárðarbunga prevented us to calculate correlations between the Brewers and OMI during these two volcanoes, as described in the text.

Comment 17: "13.16-18. This statement is based on only the Kasatochi results and does not hold for 4 of the 5 eruptions studied, thus the statement either has to be removed from the conclusions or dampened considerably by pointing out all the other times when no correlation or a poor correlation was found".

Answer to comment 17: The statement has been removed and the new text now reads: "The Kasatochi eruption provided a formidable example for a volcanic SO2 plume to be observed not only by the ground based instruments, but from space-borne as well (OMI and GOME-2). Relative to the undisturbed period before Kasatochi the amplitude of the signal is 2 DU for GOME-2 and 1.5 DU for OMI. The results for the other volcanic eruptions are similar for the Brewer network, but unfortunately because of the sparsity of satellite overpassing the Brewer stations the satellite data concur with those from the Brewers only in Kasatochi".

Reply to Reviewer #2

The authors are indebted to Reviewer #2 for his valuable comments which have all been taken into account and appropriate revisions have been done as follows:

__**Answers to main comments**__

Comment 1: "The measurement capability of Brewer instruments should be better explained. Since the paper focuses on the detection of small SO_2 signals, the methodology to derive SO_2 total content should be summarized in the paper itself. An assessment of the mean SO_2 values generally provided by Brewer instruments should be provided".

Answer to comment 1: The summary of the methodology to determine the SO_2 column has been added in section 2.1. The requested assessment emerges from our answers to comments 2 and 3 below as well as in the literature by the papers of Fioletov et al. (1998, 2016) which are referred to in the text.

Comment 2: "As optical instruments, the Brewer measurements can be perturbed by ash present in the volcanic plumes. This issue should be addressed in the article".

Answer to comment 2: We have added a relevant comment in section 2.1, in which it is shown that the presence of volcanic ash is not expected to perturb the SO_2 measurements, this addition reads as follows:
"Finally, we need to point out that perturbations by ash present in the volcanic plumes have been shown not to affect the Brewer SO_2 measurements. This is based on the result of Pappalardo et al., 2013 paper based on EARLINET observations following the Eyjafjallajökull eruption in which they found that the Ångström exponent of the volcanic ash optical depth is close to zero. This indicates that the effect of ash in the UV and visible region on the aerosol extinction is almost independent from wavelength. The Brewer SO_2 measurements taken in a narrow wavelength band in the UV are therefore not expected to be influenced by the presence of volcanic ash considering the weights already applied in the operational Brewer algorithm".

Pappalardo, G., Mona, L., D'Amico, G., et al.: Four-dimensional distribution of the 2010 Eyjafjallajökull volcanic cloud over Europe observed by EARLINET, Atmos. Chem. Phys., 13, 4429-4450, doi:10.5194/acp-13-4429-2013, 2013.

Comment 3: "For readers not familiar with total SO_2 measurements by Brewer spectrometers, it is rather intriguing to see negative total SO_2 values. So it would be worth explaining why such negative values have to be considered in the general Brewer (and satellite) retrieval".

Answer to comment 3: The following text has been added in section 2.1: "From the above described operational Brewer algorithm it is evident that the estimation of columnar SO2 is the result of the difference between two columnar terms (O3 + SO2) and O3. Both terms have uncertainties (weighting functions, calibrations, random errors, systematic errors). Systematic negative values could be the result of a systematic offset in the measurements that can be related to the calibration of the instrument (usually optimized only for the ozone measurements). Randomly varying positive and negative values around zero, suggest that the signal of SO2 is small (and thus the difference of two terms should be close to zero) but since both terms have uncertainties, negative values are possible indicating that the amount of SO2 in the atmosphere is below the detection limit of the instrument and could be considered as noise. In this work we have repeated our analysis excluding the negative values and the results remained the same i.e. a positive increase after a major volcanic eruption was confirmed as described in the following sections".

Comment 4: "Two lagrangian models are used for the analysis: FLEXPART and HYSPLIT. An explanation is needed on why two different models need to be used (paragraph 2.3)".

Answer to comment 4: Both HYSPLIT and FLEXPART are well established modelling tools and both are widely used in relevant studies. As stated in the text we use FLEXPART-WRF for the dispersion simulations. FLEXPART-WRF is driven by WRF 1-hourly data at 45×45 km and the higher spatial and temporal resolution of meteorological fields allows a more detailed representation of the volcanic plume dispersion but have significant higher computational time. To overcome this computational cost problem we use HYSPLIT for the back-trajectories calculations. HYSPLIT is driven by lower temporal and spatial resolution meteorological fields, specifically with the GDAS 3-hourly meteorology at 1°×1° resolution (see revised paragraph 2.3).

Comment 5: "In the case of the Bardarbunga volcano, the FLEXPART model has been used to simulate SO_2 levels in air masses sampled at Hohenpeissenberg station. But there is no detail on the simulation and on the initial emitted SO_2 levels".

Answer to comment 5: We thank the reviewer for this notice. The following text is now added in section 3.1: "The simulation period is 18-26 September 2014. We assume a constant SO2 release rate of 119 kilotons per day as reported by Gíslason et al. (2015) from near the source SO2 measurements during the first weeks of the eruption. Similar emission rates are also suggested by Schmidt et al. (2015) through comparisons between NAME simulations (UK Met Office's Numerical Atmospheric-dispersion Modelling Environment) and OMI satellite retrievals. The emission height is set between 0 and 3500 m above ground level, consistent throughout the simulation period".

Schmidt, A., Leadbetter, S., Theys, N., Carboni, E., Witham, C. S., Stevenson, J. A., Birch, C. E., Thordarson, T., Turnock, S., Barsotti, S, Delaney, L., Feng, W., Grainger, R. G.,Hort, M. C., Höskuldsson, A., Ialongo, I., Ilyinskaya, E., Jóhannsson, T., Kenny, P., Mather, T. A., Richards N. A. D., and Shepherd, J.: Satellite detection, long-range transport, and air quality impacts of volcanic sulfur dioxide from the 2014-2015 flood lava eruption at Bárðarbunga (Iceland), J. Geophys. Res. Atmos., 120, 9739-9757, doi:10.1002/2015JD023638, 2015.

Gíslason, S. R., Stefánsdóttir, G., Pfeffer, M. A., Barsotti, S., Jóhannsson, Th., Galeczka, I., Bali, E., Sigmarsson, O., Stefánsson, A., Keller, N. S., Sigurdsson, Á., Bergsson, B., Galle, B., Jacobo, V. C., Arellano, S., Aiuppa, A., Jónasdóttir, E. B., Eiríksdóttir, E. S., Jakobsson, S., Guðfinnsson, G. H., Halldórsson, S. A., Gunnarsson, H., Haddadi, B., Jónsdóttir, I., Thordarson, Th., Riishuus, M., Högnadóttir, Th., Dürig, T., Pedersen, G. B. M., Höskuldsson, Á., Gudmundsson, M.T.: Environmental pressure from the 2014-15 eruption of Bárðarbunga volcano, Iceland, Geochem. Persp. Let., 1, 84-93, 2015.

Comment 6: “For the same volcano, it is not completely clear that the elevated SO_2 levels detected by ground stations correspond to the volcanic plume. Also a better explanation should be given on why the plume is not seen in OMI and GOME 2 measurements shown in Figure 5. The case for the detection of this volcanic plume by the satellite instruments over Europe and for the attribution of increased SO_2 levels from these measurements (page 8) is not completely made”.

Answer to comment 6: We would like to point out that the fact that the elevated SO_2 levels detected by ground stations (Brewer network) corresponds to the volcanic SO_2 plume was confirmed by performing the back trajectories analysis with the HYSPLIT dispersion model as well as from the FLEXPART and MACC model simulations. Additionally, the Reviewer #2 correctly points out that the plume is not seen in OMI and GOME-2 measurements shown in Figure 5 (new Figure 4). We would like to note that we have carefully revisited the OMI and GOME-2 data sets and found out that during the most perturbed period following the eruptions of Bárðarbunga (21-22 September 2014) the satellite overpasses were so sparse that the daily average was not corresponding to the Brewer network sample. For instance and following Bárðarbunga, there were many days where we had only one or two OMI measurements following the eruption, obviously not representing the 19 Brewer instruments in Europe. To temper our past conclusions we have applied a criterion according to which a daily average from either OMI or GOME-2 should be calculated if and only if more than half of the individual overpasses had data at each day. As can be seen from the revised figures 4, OMI results are missing for not meeting this criterion. Also GOME-2 results are missing from the figure during the peak period (21-22/9/2014) for not passing this criterion.

In spite of the sparsity of OMI observations post Bárðarbunga, it was thought that they could still be used as SO_2 assimilated field in the SO_2 analyses and forecasts produced with the MACC (Monitoring Atmospheric Composition and Climate) system (http://atmosphere.copernicus.eu/). This near-real-time forecasting system assimilates satellite observations to constrain modelling forecasts (Inness et al., 2015; Flemming et al., 2015). The OMI instrument on board the AURA satellite provided information about concentrations of volcanic SO_2 emitted by the Icelandic Bárðarbunga volcano on 20 September; these observations were assimilated in 2014 by the MACC model in cases of volcanic eruptions, i.e. when OMI values exceeded 5 DU. As shown by the chart of total column SO_2 obtained from http://atmosphere.copernicus.eu/ (Figure 6), the subsequent forecasts then captured the transport of this plume of volcanic SO_2 southward spreading over the continent on 21 and 22 September. The plume stretched all the way from Finland through Poland, Germany and France, to southern England. A parallel forecast, for which no OMI data were used (Fig. 6, right), did not show any elevated SO_2 values, confirming that ‘normal’ emissions of SO_2 (including shipping and industrial activities) could not explain the observed situation. All the above are described in the revised text.

Comment 7: “The fact that the 2011 Grimsvötn volcanic plume was not detected by the European Brewer instrument does not bring much to the article. This paragraph should be removed”.

Answer to comment 7: The paragraph for Grimsvötn and its figures have been removed.

Comment 8: "Again for the Eyjafjallajökull volcano, OMI and GOME 2 do not seem to detect the SO_2 signal. An explanation is needed on the lack of detection by satellite instruments. Also, the left panel of Figure 16 is redundant with the right panel".

Answer to comment 8: For the case of Eyjafjallajökull, OMI and GOME-2 do not seem to detect the SO_2 signal because the satellite data were sparse, particularly OMI.
To avoid confusion the left panel of Fig. 16 has been removed. Please note that the new figure for Kasatochi is now Fig. 13.

Comment 9: "2008 Kasatochi case: it is not clear from the article why the plume is not detected in Taiwan by the satellite instruments, contrary to the observations in Europe and North America. This issue should be addressed".

Answer to comment 9: During the revision of the manuscript we analysed back trajectories from Taiwan for the days of elevated SO_2 observed by the Brewer, something that has been overlooked in the first version of the paper. The analysis showed that the air masses did not originate from Kasatochi. To avoid confusion we have removed Taiwan from the figure of Kasatochi (see new Figure 13).

Comment 10: "The conclusion should better summarize in which general conditions (SO_2 levels, time after eruption) Brewer instruments can be useful for the detection of SO_2 volcanic plumes. The article is qualitative in general and such a summary would provide a quantified assessment of the measurements capability of Brewer instruments with respect to SO_2 measurements. Comparison with OMI and GOME 2 measurements capacity in similar cases would be useful. It would be also worth mentioning why IASI and AIRS measurements are not included in the analysis".

Answer to comment 10: The concluding section has been fully revised in the new manuscript taking into consideration all the above useful comments. The second paragraph in the Conclusion has been revised and reads as follows: "From the results discussed in section 3 some general remarks can be put forward concerning SO_2 levels and detection time after the eruption. Starting with the Kasatochi eruption, as it appears from Figure 13, the plume can be detected 4 days after the eruption over Canada and the US and about 7 days over Europe with an average amplitude on the order of 2 DU compared to the unperturbed ten day pre-volcanic period (baseline). All estimates are based obviously on measurements taken under the plume. The Kasatochi eruption provided a formidable example for a volcanic SO_2 plume to be observed not only by the ground based instruments, but from space-borne as well (OMI and GOME-2). Relative to the undisturbed period before Kasatochi the amplitude of the signal is 2 DU for GOME-2 and 1.5 DU for OMI. The results for the other volcanic eruptions are similar for the Brewer network, but unfortunately because of the sparsity of satellite overpassing the Brewer stations the satellite data concur with those from the Brewers only in Kasatochi. Based on the above discussion it appears that currently no single network can independently and fully monitor the evolution of volcanic SO_2 plumes. Among a few reasons are lack of measurements during peak values, complications from meteorological events, ejection heights and exposure conditions. The evidence presented here points that combination of observations from various instruments, aided by chemical transport models and operated in synergy could address such a complex issue".

Additionally, we want to point out that we did not consider in this paper SO_2 measurements from IASI and AIRS since both instruments are IR spectroradiometers, while OMI and GOME-2 data are based on UVB/Vis spectroradiometers whose retrieval algorithms rely on the differential optical absorption in the UV band which is also the case with the Brewer instrument. A Brewer-IASI or Brewer-AIRS comparison would also have to consider differences in the spectroscopy and algorithm concept and thus would require further analysis which is beyond the scope of this paper.

Answers to minor comments

Comment 11: "In general, figures' legends should be more informative, with the description of the various plots and the name of the volcano case to which the figure refer (when SO_2 levels are plotted)".

Answer to 11: The figures' legends have been re-written to be more informative as suggested by the reviewer.

Comment 12: "Figure 7: can the authors comment on the spot of elevated SO_2 observed between Italy and Greece?"

Answer to 12: The spot of elevated SO_2 between Italy and Greece is related to the Etna volcano and is a result of using continuous natural SO_2 emissions that might be too high in the MACC model.

Further additions to the manuscript

Three more stations have been added, namely Regina and Goose Bay in Canada and Mauna Loa in the US. Two more co-authors have been added, Vitali Fioletov and Irina Petropavlovskikh, who provided the SO_2 column data for these additional stations.

Detecting volcanic sulfur dioxide plumes in the Northern Hemisphere using the Brewer spectrophotometers, other networks, and satellite observations

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Abstract. This study examines the adequacy of the existing Brewer network to supplement other networks from ground and space to detect SO_2 plumes of volcanic origin. It was found that large volcanic eruptions of the last decade in the Northern Hemisphere have a positive columnar SO_2 signal seen by the Brewer instruments located under the plume. It is shown that a few days after the eruption the Brewer instrument is capable of detecting significant columnar SO_2 increases on the average exceeding 2 DU relative to a considered as unperturbed pre-volcanic baseline 10-day-period, with a mean close to zero and $\sigma = 0.46$, as calculated from the 33 Brewer instruments under study. Intercomparisons with independent measurements from ground and space as well as theoretical calculations corroborate the capability of the Brewer network to detect volcanic plumes. For instance, the comparison with OMI and GOME-2 SO_2 space-borne retrievals shows statistically significant agreement between the Brewer network data and the collocated satellite overpasses in the case of Kasatochi eruption. Unfortunately, due to sparsity of satellite data the significant positive departures seen in the Brewer and other ground networks following Eyjafjallajökull, Bárðarbunga and Nabro eruptions could not be statistically confirmed by the data from satellite overpasses. ~~This paper demonstrates that SO_2 columnar amounts have significantly increased following the five largest volcanic eruptions of the past decade in the Northern Hemisphere. A strong positive signal was detected by all the existing networks either ground based (Brewer, EARLINET, AirBase) or from satellites (OMI, GOME-2). The study particularly examines the adequacy of the existing Brewer network to detect SO_2 plumes of volcanic origin in comparison to other networks and satellite platforms. The comparison with OMI and GOME-2 SO_2 space-borne retrievals shows statistically significant agreement between the Brewer network data and the collocated satellite overpasses. It is shown that the Brewer instrument is capable of detecting significant columnar SO_2 increases following large volcanic eruptions, when~~

~~SO_2 levels rise well above the instrumental noise of daily observations, estimated to be of the order of 2 DU.~~ A model exercise from the MACC (Monitoring Atmospheric Composition and Climate) project shows that the large increases of SO_2 over Europe following the Bárðarbunga eruption in Iceland were not caused by local pollution sources or ship emissions but are clearly linked to the volcanic eruption. Sulfur dioxide positive departures in Europe following Bárðarbunga could be traced by other networks from the free troposphere down to the surface (AirBase and EARLINET). We propose that by combining Brewer data with that from other networks and satellites, a useful tool aided by trajectory analyses and modeling could be created which can be used also to forecast high SO_2 values both at ground level and in air flight corridors following future eruptions.

1 Introduction

Volcanic eruptions are an important source of natural emissions of sulfur dioxide (SO_2) into the troposphere and the stratosphere. Ash particles and gases injected into the atmosphere by large volcanic eruptions can affect solar radiation and climate (e.g. Robock, 2000), air quality (e.g. Schmidt et al., 2015) and may also impact local environments (e.g. Durant et al., 2010). Volcanic emissions (e.g ash and SO_2) can reach different heights in the atmosphere and can be transported in different directions (e.g. Prata et al., 2010). Thomas and Prata (2011) have shown that the eruption can be divided into an initial ash rich phase, a lower intensity middle phase and a final phase where considerably greater quantities both ash and SO_2 are released which in the case of ash can result even to air travel disruptions (e.g. Flentje et al., 2010). These effects make the ash and SO_2 in volcanic plumes important parameters to be studied, monitored and forecasted on small and larger spatial scales. Our study focuses on volcanic columnar SO_2 amounts because of the existence of the rather continuous set of direct sun measurements with the Brewer network.

Measurements of SO_2 are important for tracking and assessing impacts of emissions from pollution sources and in quantifying natural SO_2 emissions by volcanoes. Pollution sources typically result in a few Dobson Units (DU, 1 DU = $2.69 \cdot 10^{26}$ molec•km^{-2}) increases of column SO_2 amounts unless observations are made near a source. The Brewer network is useful for plume tracking because it can track SO_2 columnar amounts from a large number of stations and wide geographical extent.~~Brewer instruments are useful for plume tracking because they measure columnar amounts and because the network is quite extended.~~ The primary application of the ground-based Brewer spectrophotometer is to measure ozone by using UV spectrophotometry. Direct sunlight intensities are measured at five wavelengths (between 306 and 320 nm; see also Sect. 2.1) to simultaneously calculate ozone and SO_2 column integrals (Kerr et al., 1980). These instruments have been used extensively to monitor stratospheric ozone (e.g. WMO Scientific Assessment of Ozone Depletion~~ozone depletion~~ reports 2011, 2014) and have a long history of studying atmospheric SO_2 columns (e.g. De Backer and De Muer, 1991; Bais et al., 1993; Fioletov et al., 1998; Zerefos et al., 2000; Zerefos et al., 2009; Ialongo et al., 2015). Ground-based measurements of atmospheric SO_2 using the Brewer instrument have played an important role in the development and validation of satellite-based SO_2 measurements (Schaefer et al., 1997; Spinei et al., 2010; Rix et al., 2012; Ialongo et al., 2015) used primarily for detecting and tracking volcanic emissions. Since the Brewer instruments are typically used as stationary ground-based monitoring sites, a volcanic plume of SO_2 must pass over the site if useful data are to be obtained. Validation of satellite measurements by the Brewer instrument also requires that a satellite overpass is available when the plume is over ~~or nearby~~ the ground based site (Kerr, 2010).

There have been various initiatives during recent years that used satellite measurements of SO_2 to monitor volcanic eruptions in support of aviation safety, e.g. ESA's Support to Aviation Control Service (SACS) (Brenot et al., 2014). These initiatives together with modelling forecasting tools provide valuable information to the established Volcanic Ash Advisory Centers (VAAC). Satellite SO_2 data have been available in the past from various satellite instruments (e.g. GOME, SCIAMACHY). Currently operational data are available from UV measurements (e.g. GOME-2 (Global Ozone Monitoring Experiment-2), OMI (Ozone Monitoring Instrument) and OMPS (Ozone Mapping Profiler Suite)) and from infrared measurements (e.g. IASI (Infrared Atmospheric Sounding Interferometer) and AIRS (Atmospheric Infrared Sounder)). ~~There have been various initiatives during recent years that used satellite measurements of SO_2 to monitor volcanic eruptions focusing mostly on aviation, e.g. ESA's Support to Aviation Control Service (SACS) (Brenot et al., 2014). These initiatives together with modeling forecasting tools provide valuable information to the established Volcanic Ash Advisory Centers (VAAC). Satellite SO_2 data have been available in the past from various instruments (e.g. GOME, SCIAMACHY) but currently data are operationally available from GOME-2, OMI and OMPS based on UV measurements and IASI and AIRS based on infrared measurements.~~

In the present work we investigate the efficiency of the existing Brewer network in the Northern Hemisphere to detect volcanic SO_2 plumes during the past decade. The main focus is to show the sensitivity of the Brewer network in detecting SO_2 plumes of volcanic origin in synergy with other ground based observations, satellite data and dynamic transport calculations. The Brewer spectroradiometric measurements are compared to collocated satellite measurements from OMI and GOME-2 as described in the next paragraph. This paper did not include analyses of the SO_2 measurements from IASI and AIRS since both instruments are IR spectroradiometers. We compared Brewer measurements against the OMI and GOME-2 data that are derived using information from differential optical absorption in the UV spectrum, which is also at the base of the Brewer measurement methodology. In the case of Brewer-IASI or Brewer-AIRS comparison we would also have to consider differences in the spectroscopy and the corresponding retrieval algorithm concepts, which would require further analysis which is beyond the scope of this paper.

Table 1 lists in chronological order all major volcanic eruptions in the Northern Hemisphere between 2005-2015 with volcanic explosivity scale index (VEI) of at least 4 (Newhall and Self, 1982; Robock et al., 2000; Zerefos et al., 2014). The study also provides a separate analysis for the Bárðarbunga eruption, which although not rated 4 has been already studied with the Brewer at Sodankylä by Ialongo et al. (2015).

As seen from Table 1, chronologically, the first case was the volcanic eruption at Mount Okmok, Alaska (53.43^o N, 168.13^o W, 1073 m above sea level (asl), 12 July 2008, Prata et al., 2010) followed by the Kasatochi eruption, Alaska (52.17^o N, 175.51^o W, 300 m asl, 7-8 August 2008, e.g., Kristiansen et al., 2010; Krotkov et al., 2010; Waythomas et al., 2010) which was detected over large areas of the Northern Hemisphere. Okmok and Kasatochi volcanoes in Alaska erupted a short time span of less than a month and therefore we decided to study the evolution of the Brewer SO_2 columnar measurements following the latter volcanic eruption (Kasatochi). The third eruption took place at Sarychev in Russia (48.1^{o} N, 153.2^{o} E, 1496 m asl, 12-17 June 2009, Haywood et al., 2010). The evolution of the SO_2 volcanic plume from Sarychev was mostly observed over the North Pacific, North America and North Atlantic (Haywood et al., 2010). There was only one North American Brewer station (Saturna Island) in the path of the plume from Sarychev eruption. The record shows SO_2 columns of 8.6 DU detected on 19 June 2009 and 3.7 DU on 20 June 2009. This volcanic eruption is not investigated any further in this paper. The next eruption on the list, Eyjafjallajökull in Iceland (63.63^{o} N, 19.62^{o} W, 1666 m asl, from 14 April to 23 May 2010), resulted in interruption of the air traffic over NW Europe (e.g. Flemming and Inness, 2013). The fifth eruption Grímsvötn 2011 (64.42^{o} N, 17.33^{o} W, 1725 m asl, 21 May 2011) was studied by Flemming and Inness (2013), and by Moxnes et al. (2014). This eruption provided an interesting example of a clear separation of the volcanic SO_2 plume (transported mostly northwestward) while the fine ash was transported mostly southeastward. Unfortunately the volcanic plume did not overpass any Brewer station and therefore we do not include any results post Grímsvötn eruption. The sixth eruption recorded features the Nabro in Africa (13.37^{o} N, 41.70^{o} E, 2218 m asl) that occurred on 12-13 June 2011 (e.g., Bourassa et al., 2012; Sawamura et al., 2012; Clarisse et al., 2014). We present here a case study that described detection of the Nabro volcanic SO_2 plume over ground based stations. The plume was clearly detected by the Brewer instrument over Izaña (and poorly from space), then over Taiwan by both Brewer and satellite instruments, and finally at Mauna Loa, Hawaii (mostly by the Brewer instrument). The seventh eruption was Tolbachik, Russia (55.83^{o} N, 160.33^{o} E, 3.611 m asl) on 27 November 2012 (e.g. Telling et al., 2015). As in the case of Grímsvötn, the plume has not passed over any Brewer station that was verified by trajectory analysis. The next eruption on the list is the volcanic eruption from Bárðarbunga, Iceland (64.64^{o} N, 17.56^{o} W, 2005 m asl) that was observed between 31 August 2014 and 28 February 2015 (e.g. Schmidt et al., 2015). This last eruption, although not yet rated on the VEI scale, has been extensively studied in view of the observed increased SO_2 concentrations that have been observed all the way through troposphere and reaching down to the surface in Europe (Ialongo et al., 2015; Schmidt et al., 2015). ~~Five cases of high SO_2 from volcanic eruptions listed in Table 1, and shown in Figure 1 over Iceland, with distinct columnar SO_2 characteristics and plume trajectories, are compared in this study. These include large volcanic eruptions that have occurred in the Northern Hemisphere in the past decade (2005-2015) measuring in the volcanic explosivity scale index at least 4 (VEI; Newhall and Self, 1982; Robock et al., 2000; Zerefos et al., 2014). Although the area of study is the Northern Hemisphere, we note here that Europe has a dense Brewer network which is operating with accessible long term columnar SO_2 data. We also note here that there were two more volcanic eruptions rated 4 during the period under study, namely, Mount Okmok, Alaska, (53.43^{o}N, 168.13^{o}W, 1073 m above sea level (asl), 12 July 2008, Prata et al., 2010) and Sarychev, Russia (48.1^{o}N, 153.2^{o}E, 1496 m asl, 12-17 June 2009, Haywood et al., 2010). Okmok and Kasatochi volcanoes in Alaska erupted within less than a month and therefore we decided to study the evolution of the Brewer SO_2 columnar measurements following the latest volcanic eruption (Kasatochi). The evolution of the SO_2 volcanic plume from Sarychev was mostly observed over the North Pacific, North America and North Atlantic (Haywood et al., 2010). Unfortunately there was only one Brewer station under the plume over North America following Sarychev, measuring SO_2 columns of 8.6 DU on 19 June 2009 and 3.7 DU on 20 June 2009 (Saturna Island, not shown here), so this volcanic eruption was not investigated any further here.~~

~~As seen from Table 1, chronologically, the first case is the Kasatochi eruption in Alaska (52.17°N, 175.51°W), 300 m asl, which erupted on 7-8 August 2008, (e.g., Kristiansen et al., 2010; Waythomas et al., 2010) and was detected over large areas of the Northern Hemisphere. The next eruption is Eyjafjallajökull in 2010 (63.63°N, 19.62°W, 1666 m asl, from 14 April to 23 May 2010), responsible for the interruption of air traffic over NW Europe (e.g. Flemming and Inness, 2013). The third is Grímsvötn 2011 eruption (64.42°N, 17.33°W, 1725 m asl, 21 May 2011), studied also by Flemming and Inness (2013) and by Moxnes et al. (2014). This is an interesting example of a clear separation of the volcanic SO_2 plume (transported mostly northwestward) and the fine ash (transported mostly southeastward). The fourth is the Nabro eruption in Africa (13.37°N, 41.70°E, 2218 m asl, 12-13 June 2011, e.g., Bourassa et al., 2012; Clarisse et al., 2014). Here we present a case where the volcanic SO_2 plume from this eruption is detected over Izaña mostly by the Brewer instrument (and poorly from space) but over Taiwan by both. The fifth is the Bárðarbunga eruption (64.64°N, 17.56°W, 2005 m asl, between 31 August 2014 and 28 February 2015, e.g., Schmidt et al., 2015) after which increased SO_2 concentrations have been observed down to ground level in Europe.~~

The capability of the Brewer network to measure columnar SO_2 amounts above the local air pollution levels is also presented and discussed. The qualitative evidence that the plume can be detected in many single cases by the Brewer network has been quantitatively tested by calculating correlation coefficients with collocated satellite data. Only in the case of Kasatochi 2008 eruption it was possible to test the sensitivity of SO_2 abundance measured by the Brewers and from space.~~We have selected the case of Kasatochi 2008 eruption because of its importance both in intensity, duration and its large scale spreading over the majority of the Brewer stations.~~ Correlations between the Brewer and collocated satellite SO_2 data from the Aura OMI and GOME-2 are presented in section 3 where the correlation coefficients were found to be statistically significant at a confidence level of 99%. For the other eruptions unfortunately due to the sparsity of satellite data no firm conclusions can be drawn as discussed in section 3.

The paper is structured in the following ~~Sections:~~order, Section 2 describes the data sources and the methods of analysis of the columnar SO_2 measurements by the Brewer spectrophotometers (hereinafter simply referred to as the “Brewers”). Section 3 presents the analysis of the Brewer measurements during four of the~~the five~~ volcanic eruptions listed in Table 1, along with satellite data and dynamic volcanic plume transport simulations. The conclusions are provided in Section 4.

2 Data and methods

2.1 Ground based data

Sulfur dioxide~~SO_2~~ in the atmosphere can be measured from ground-based instruments, by instrumentation onboard the spacecraft and can be estimated with help of~~calculated with~~ models. The Brewer is an automated, diffraction-grating spectrophotometer that provides observations of the sun’s intensity in the near UV range. The spectrophotometer measures the intensity of light in the ultraviolet absorption spectrum of ozone at five wavelengths (306.3 nm, 310.1 nm, 313.5 nm, 316.8 nm and 320.1 nm) with a resolution of 0.6 nm. These data are used to derive the total ozone column (Kerr et al., 1980). Because sulfur dioxide has strong and variable absorption in this spectral region, the Brewer spectrophotometer has additionally been proposed to derive SO_2 columns~~is additionally used to derive the SO_2 column~~ (Kerr et al., 1980). About two hundred Brewer spectrophotometers around the world contribute high-precision ozone data to the global ozone monitoring network (Kumharn et al., 2012). The existing Brewer network also delivers frequent SO_2 columnar measurements as well, which can be used for analyses, but with caution.~~The existing Brewer network could deliver frequent SO_2 measurements as well, but the Brewer instruments are less able to accurately provide SO_2 measurements.~~ This is because the signal to noise ratio for the SO_2 absorption is usually quite low and therefore well calibrated instruments are required to monitor nominal SO_2 columnar amounts~~levels~~ (Koukouli et al., 2014). Details on the method with which SO_2 is measured ~~with~~ by the Brewer spectrophotometer can be found in Kerr et al. (1980; 1985; 1988) and De Backer and De Muer (1991). According to Fioletov et al. (2016) the uncertainty of the Brewer direct sun (DS) SO_2 measurements is about 1 DU and is typically insufficient for air quality applications.~~The uncertainty of the Brewer direct sun (DS) SO_2 measurements is about 1-2 DU (1 DU is equal to 2.69 x 10^{16} molecules/cm^2) and is typically insufficient for air quality applications (Fioletov et al., 2016).~~ A more accurate method (with an uncertainty as low as 0.13 DU) based on Brewer "group-scan" spectral direct sun radiation measurements at 45 wavelengths from 306 to 324 nm was developed (Kerr, 2002), but not ~~widely~~ implemented for routine operations due to its complexity (Fioletov et al., 2016). Although the Brewer instrument has difficulties in detecting low columnar SO_2 concentrations, in extreme cases, such as volcanic eruptions, the SO_2 levels typically rise well above the instrumental noise and can be identified with the Brewer instrument as shown in this paper and in Fioletov et al. (1998). ~~However, the high uncertainty of the SO_2 column measurement of the Brewer has not been investigated with the same attention as ozone, resulting in larger uncertainties due to the calibration itself, the transfer from Brewer to Brewer, and the cross sections themselves.~~

Before proceeding to the analysis of Brewer measurements, the methodology to derive columnar SO_2 is first presented. To determine ozone and SO_2 column amounts, the measured raw photon counts at the five operational channels in the Brewer instrument are converted to radiation intensity. The Beer-Lambert absorption law is applied at each wavelength λ, and the measured intensity of direct sunlight is given by the formula:

$$\log I_\lambda = \log I_{0\lambda} - \beta_\lambda \mu_R - \delta_\lambda \mu_p - \alpha_\lambda O_3 \mu - \sigma_\lambda SO_2 \mu \qquad (1)$$

where $\boldsymbol{I_\lambda}$ is the measured radiation intensity at wavelength λ, $\boldsymbol{I_{0\lambda}}$ is the measured extra-terrestrial spectrally resolved intensity at λ, $\boldsymbol{\beta_\lambda}$ is the Rayleigh scattering coefficient at λ, $\boldsymbol{\delta_\lambda}$ is the particulate scattering coefficient at λ, $\boldsymbol{\alpha_\lambda}$ is the ozone absorption coefficient (cm^2/molecules) at λ, $\boldsymbol{O_3}$ is the total ozone column (molecules/cm^2), $\boldsymbol{\sigma_\lambda}$ is the SO_2 absorption coefficient at λ, $\boldsymbol{SO_2}$ is the column amount of sulfur dioxide, $\boldsymbol{\mu_R}$, $\boldsymbol{\mu_p}$ and $\boldsymbol{\mu}$ are the optical path lengths (air masses) corresponding to the effective heights of molecules, particles, and ozone respectively.

According to the Brewer retrieval algorithm, the following ratios are formed:

$$F = F_0 - \Delta\beta \mu_R - \Delta\alpha O_3 \mu \qquad (2)$$

and

$$F' = F'_0 - \Delta\beta'\mu_R - \Delta\alpha' O_3\mu - \Delta\sigma' SO_2\mu \qquad (3)$$

where $\boldsymbol{F}$ is the weighted ratio of direct sun measurements at 4 (or 6 for double Brewer) spectral channels, $F = logI_2 - 0.5\, logI_3 - 2.2\, logI_4 + 1.7\, logI_5$, $\boldsymbol{F_0}$, $\boldsymbol{\Delta\beta}$, and $\boldsymbol{\Delta\alpha}$ are the same linear combinations for $\mathbf{log}\boldsymbol{I_{0\lambda}}$, $\boldsymbol{\beta_\lambda}$, and $\boldsymbol{\alpha_\lambda}$. The $\boldsymbol{F'}$ is the SO_2 ratio, $F' = logI_1 - 4.2\, logI_4 + 3.2\, logI_5$ and $\boldsymbol{F_0'}$, $\boldsymbol{\Delta\beta'}$, $\boldsymbol{\Delta\alpha'}$ and $\boldsymbol{\Delta\sigma'}$ the corresponding linear combinations for $\mathbf{log}\boldsymbol{I_{0\lambda}}$, $\boldsymbol{\beta_\lambda}$, $\boldsymbol{\alpha_\lambda}$, $\boldsymbol{\sigma_\lambda}$. Both of these functions have weights which eliminate the effects of particulate scattering, while the function $\boldsymbol{F}$ is weighted to remove SO_2 absorption effects as well. The extra-terrestrial constants $\boldsymbol{F_0}$ and $\boldsymbol{F_0'}$ are determined from a long series of intercomparison measurements as well as zero air mass ($\boldsymbol{\mu}$) extrapolations.

The total ozone column is determined by the formula

$$O_3 = \frac{F_0 - F - \Delta\beta\mu_R}{\Delta\alpha\,\mu} \qquad (4)$$

and the SO_2 by the formula

$$SO_2 = \frac{1}{A}\left(\frac{F'_0 - F' - \Delta\beta'\mu_R}{\Delta\alpha'\,\mu} - O_3\right) \qquad (5)$$

where $\boldsymbol{A}$ is the ratio of the SO_2 absorption coefficient to the O_3 absorption coefficient, $\boldsymbol{A}$ = 2.44.

From the above described operational Brewer algorithm it is evident that the estimation of columnar SO_2 is the result of the difference between two columnar terms (O_3 + SO_2) and O_3. Both terms have uncertainties (weighting functions, calibrations, random errors, systematic errors). Systematic negative values could be the result of a systematic offset in the measurements that can be related to the calibration of the instrument (usually optimized only for the ozone measurements). Randomly varying positive and negative values around zero, suggest that the signal of SO_2 is small (and thus the difference of two terms should be close to zero) but since both terms have uncertainties, negative values are possible indicating that the amount of SO_2 in the atmosphere is below the detection limit of the instrument and could be considered as noise. In this work we have repeated our analysis excluding the negative values and the results remained the same i.e. a positive increase after a major volcanic eruption was confirmed as described in the following sections. Finally, we need to point out that perturbations by ash present in the volcanic plumes have been shown not to affect the Brewer SO_2 measurements. This is based on the result of Pappalardo et al., 2013 paper based on EARLINET observations following the Eyjafjallajökull eruption in which they found that the Ångström exponent of the volcanic ash optical depth is close to zero. This indicates that the effect of ash in the UV and visible region on the aerosol extinction is almost independent from wavelength. The Brewer SO_2 measurements taken in a narrow wavelength band in the UV are therefore not expected to be influenced by the presence of volcanic ash considering the weights already applied in the operational Brewer algorithm.

In this study we analysed twenty three stations located in ~~the European Union~~Europe, ~~four~~ six Brewer stations in Canada, ~~one~~ two in the USA and one in Taiwan, whose geographical positions are shown in Figure 1~~2~~. SO_2 measurements were averaged over a large number of instruments and datasets during periods following volcanic eruptions. Random errors in the measurements of individual Brewer stations are reduced significantly by the averaging processes to calculate regional means.

Daily SO_2 columns at Churchill, Goose, Edmonton, Regina, Saturna Island~~,~~ and Toronto in Canada, ~~and~~ Taipei in Taiwan, Boulder and Mauna Loa in the US were obtained from the World Ozone and Ultraviolet Radiation Data Centre (WOUDC; http://www.woudc.org/)~~. SO_2 columns at Niwot Ridge, USA, were available for download from~~ and the NOAA-EPA Brewer Spectrophotometer UV and Ozone Network (NEUBrew; http://www.esrl.noaa.gov/gmd/grad/neubrew/). The data have been checked for quality assurance/quality control by the individual data providers. It is important to note the participation of the~~We mention here that~~ most of the European Brewer data providers ~~participate~~ in a recent EU COST Action (EUBREWNET, http://www.eubrewnet.org/cost1207/) programme. ~~aiming~~ Its focus is at establishing a coherent network of European Brewer Spectrophotometer monitoring stations in order to harmonise operations and develop approaches, practices and protocols to achieve consistency in quality control, quality assurance and coordinated operations.

In our analysis only direct sun (DS) measurements satisfying the following criteria have been used: A Brewer DS measurement was included in our analysis only if for every measurement cycle of 5 sets of measurements (from which also total columnar ozone is derived) the standard deviation of O_3 and SO_2 was less than 2.5 DU, the total columnar ozone was between 250 DU and 450 DU, and the solar zenith angle was less than 73.5 degrees. To exclude erratic data of SO_2 from our analysis, values exceeding $\pm 6\sigma$ of the mean of all SO_2 individual Brewer measurements were considered unrealistic and were not included in the calculations. Therefore the range of analysed values were limited to a maximum of $\pm$ 35 DU for an individual measurement (i.e. 6σ, with σ being equal to 5.8 as estimated from all available sub-daily SO_2 values). Then we calculated daily SO_2 columns at each station only if at least three individual measurements passed these criteria for each day. Brewers are useful because they provide more than one observation per day. For plumes which change rapidly, more than one observation per day would be useful, especially to complement satellites which typically have just one local overpass.

Daily sulfur dioxide (SO_2) columns were analysed in ~~five~~ four bimonthly periods, namely August-September 2008, April-May 2010, ~~May-June 2011,~~ June-July 2011 and September-October 2014, which include the volcanic eruptions of Kasatochi (2008), Eyjafjallajökull (2010), ~~Grímsvötn (2011),~~ Nabro (2011) and Bárðarbunga (2014), respectively. For the case of Kasatochi, Eyjafjallajökull, ~~Grímsvötn~~ and Bárðarbunga we analysed daily SO_2 columns at ~~twenty seven~~30 sites ~~located at middle latitudes~~ (listed in Table 2), while for the case of Nabro, whose impact was mostly seen over low latitudes in the N.H. (e.g., Bourassa et al., 2012), we analysed SO_2 columns at ~~two~~ three low latitude sites in the Northern Hemisphere, namely Izaña, Mauna Loa and Taipei.

Only for the case of the Bárðarbunga eruption in 2014, the columnar SO_2 measurements over Europe were also compared with measurements from ground based European stations from the European Environment Agency databases (AirBase) covering the bimonthly period September-October 2014. Only rural background stations, i.e stations in class 1-2 according to the Joly-Peuch classification methodology for the surface sulfur dioxide (Joly and Peuch, 2012), located at a distance of less than 150 km from the nearest Brewer station, were used in the analysis. A total of 7 stations in Europe (see Table 3) fulfilled the above mentioned criteria and were included in the current analysis. Observed data from the AirBase network were available in hourly resolution, from which we calculated daily surface SO_2 values. We note here that SO_2 in the troposphere over Western Europe is very low (e.g. Zerefos et al., 2009; Wild, 2012) and therefore plumes from volcanic eruptions are more easy to detect against a lower ~~noise~~ background level.

2.2 Satellite Data

The columnar SO_2 records from remote sensing spectrophotometers over Europe, Canada, USA and Taiwan were compared with space-borne measurements from a) the Ozone Monitoring Instrument (OMI) ~~on-board~~aboard the EOS-Aura (e.g. Ialongo et al., 2015) satellite and b) the Global Ozone Monitoring Experiment-2 (GOME-2) ~~on board~~aboard the MetOp-A (e.g. Rix et al., 2009). We use MetOp-A instead of MetOp-B because it covers a longer time period. Both OMI and GOME-2 satellite SO_2 data products were downloaded from the Aura Validation Data Center (AVDC) ~~(at the website~~available from: http://avdc.gsfc.nasa.gov/index.php?site=245276100~~)~~. GOME-2 level 2 overpass data have been processed with the GOME Data Processor (GDP) version 4.7. We analysed station overpass data for the various mid-latitude stations listed in Table 2 and for the low latitude stations at Mauna Loa, Izaña and Taipei. The available OMI version 1.2.0 overpass (collection 3) data analysed in this study include pixels within 50 km radius from the nearest Brewer site and is not affected by OMI row anomalies. ~~, and the~~The available GOME-2 level 2 overpass data include pixels within 100 km radius from the Brewer sites.

For the case of OMI, the SO_2 data are provided from October 2004 to the present. There are four SO_2 products: (1) the Planetary Boundary Layer SO_2 column (PBL), corresponding to a centre of mass altitude (CMA) of 0.9 km, (2) the lower tropospheric SO_2 column (TRL), corresponding to CMA of 2.5 km, (3) the middle tropospheric SO_2 column (TRM), usually produced by volcanic degassing, corresponding to CMA of 7.5 km, and (4) the upper tropospheric and stratospheric SO_2 column (STL), usually produced by explosive volcanic eruptions, corresponding to CMA of 17 km. Details on OMI SO_2 columns can be found in various studies (Levelt et al., 2006; Yang et al., 2007; Fioletov et al., 2011; McLinden et al., 2012; Fioletov et al., 2013; Li et al., 2013; Ialongo et al., 2015). In this study, we primarily made use of the product for the middle tropospheric SO_2 column (TRM) following the recommendation that the TRM retrievals should be used for volcanic degassing at all altitudes, because the PBL retrievals are restricted to optimal viewing conditions and TRL data are overestimated for high altitude emissions (>3km) (Ialongo et al., 2015). The standard deviation of TRM retrievals in background areas is reported to be about 0.3 DU in low and mid-latitudes. This is similar to the standard deviation (indicative of typical uncertainties of the measurements) that we find for the TRM retrievals in the four bimonthly periods under study. For best data quality, we used data from scenes near the centre of the OMI swath (rows 4-54) as recommended by Ialongo et al. (2015) who found that ~~, as~~ data from the edges of the swath tend to have greater noise ~~(Ialongo et al., 2015)~~.

For GOME-2, we analysed the total SO_2 columns from April 2007 to the present. The standard deviation found in our study for the GOME-2 retrievals is the order of 0.4 DU. We analysed satellite SO_2 measurements when O_3 column was between 250 and 450 DU and solar zenith angle was less than 73.5 degrees. We used SO_2 data defined as having a cloud radiance fraction (across each pixel) less than 50%, as they were found to have smaller standard deviation than all sky data. Moreover, a range of SO_2 values between -35 and 35 DU was used to screen for outliers. In cases of multiple daily data matched to the station overpass, all available measurements within a radius of 50 (100) km from the Brewer site in the case of OMI (GOME-2) are averaged.~~Again, we considered a range of SO_2 values between -35 and 35 DU. In cases when more than one overpass fulfilled these criteria for each day we took the average of all available measurements within a radius of 50 km from the Brewer site in the case of OMI and 100 km for the case of GOME-2.~~

Finally, both for the satellite and the Brewer data we have considered that during a ten-day period prior to any eruption both the surface and the satellite data sets represent a baseline reference from which subsequent departures after the eruption should be tested as to their significance. Therefore, we calculated averages and standard deviations (σ) of departures from the unperturbed pre-volcanic period, for the three studied periods of volcanic importance at each station, only if at least 25 daily values were available. The bimonthly averages for each station in the examined periods are presented in Table 4a. Table 4b shows the mean and standard error (σ/√N) of all bimonthly averages in each studied volcanic period. Averaging the departures from the pre-volcanic baseline for all Brewer stations and for all bimonthly periods gives a mean SO_2 columnar departure of 0.10 ± 0.03 DU. This estimate is on the same order of magnitude as the corresponding statistics for OMI (TRM) SO_2 column departures (0.05 ± 0.02 DU) and that measured by GOME-2 (0.09 ± 0.02 DU). The standard deviation of the bimonthly averages relative to their baselines, which was calculated from a large sample of data, was taken here as an approximation of the typical uncertainties in the columnar SO_2 measurements performed by the group of Brewers, OMI and GOME-2 instruments following volcanic eruptions.~~Finally, both for the Brewer and satellite data we calculated bimonthly averages and standard deviations (σ) for the 4 study periods of volcanic importance at each station, only if at least 25 daily averages were available in each bimonthly period. The bimonthly averages for each station in the examined periods are presented in Table 4a. Table 4b shows the mean and standard error (σ/√N) of all bimonthly averages in each period. Averaging the data from all examined Brewer stations and for all bimonthly periods gives a mean SO_2 column amounting to 0.46 ± 0.14 DU. This estimate is greater than the mean OMI (TRM) SO_2 column (-0.02 ± 0.02 DU) and that measured by GOME-2 (0.09 ± 0.02 DU). The standard deviation of the bimonthly averages, which was calculated from a large sample of data, was taken here as an approximation of the typical uncertainties in the columnar SO_2 measurements performed by the Brewers, the OMI and GOME-2 instruments.~~

2.3 Modeling tools

Dispersion of volcanic emissions is simulated with the Lagrangian transport model FLEXPART (Stohl et al., 2005; Brioude et al., 2013). The model is driven by hourly meteorological fields from the Weather Research and

Forecasting (WRF) atmospheric model (Skamarock et al., 2008) at a horizontal resolution of 45×45 km. ~~Initial~~ The initial and boundary conditions for the WRF model are taken from the National Center for Environmental Prediction (NCEP) final analysis (FNL) dataset at 1°×1° resolution. ~~and the~~The sea surface temperature (SST) is initialised from the NCEP 1°×1° analysis. The use of 1-hourly WRF meteorological fields at 45x45 km spatial resolution allows a more detailed representation of the volcanic plume dispersion but implies also a significant increase in computational time. To overcome this computational time cost, source-receptor relationships between station measurements and volcanic activity are also analysed with the use of HYSPLIT model trajectories (Stein et al., 2015) of long range transport driven by the 3-hourly meteorological dataset Global Data Assimilation System (GDAS) at a resolution of 1°×1°.~~A total of 40,000 tracer particles are assumed for each release in FLEXPART simulations. Source-receptor relationships between station measurements and volcanic activity are also analyzed with the use of HYSPLIT model (Stein et al., 2015). HYSPLIT forward and backward trajectories of long range transport are driven by the 1°×1° Global Data Assimilation System (GDAS) meteorological dataset.~~

3 Results and discussion

3.1 The 2014 Bárðarbunga case

A detailed description of the transport of Bárðarbunga plumes towards the station of Hohenpeissenberg is provided using the FLEXPART Lagrangian particle dispersion model offline coupled with the WRF_ARW atmospheric model. The simulation period is 18-26 September 2014. We assume a constant SO_2 release rate of 119 kilotons per day as reported by Gíslason et al. (2015) from near the source SO_2 measurements during the first weeks of the eruption. Similar emission rates are also suggested by Schmidt et al. (2015) through comparisons between NAME simulations (UK Met Office's Numerical Atmospheric-dispersion Modelling Environment) and OMI satellite retrievals. The emission height is set between 0 and 3500 m above ground level, consistent throughout the simulation period. The establishment of an anticyclonic flow over the British Isles on 21 September 2014 (not shown here) resulted in the separation of the volcanic SO_2 field into two distinct plumes (Figure ~~3a~~2a). On 22 September the primary plume (plume~~_~~ 1) becomes stagnant over the topographic barrier of the Alps (Figure ~~3b~~2b). The secondary plume is advected southwards by the intense northerly winds over the North Sea. ~~and the~~The two plumes overlap at about 09:00-11:00 UTC. Taking a closer look at the surface SO_2 values sampled during this event by surface air quality stations in the Netherlands, several days of enhanced SO_2 were discovered, which indicate an area of stagnation or blocking of the flow.~~Taking a closer look at the surface SO_2 in Netherlands for this event from surface air quality stations, we found several days of enhanced SO_2 indicating an area of stagnation or blocking of the flow.~~ Trajectory calculations performed at the Royal Netherlands Meteorological Institute (KNMI) correspond well to the calculations shown in Figure ~~3~~2, but also show that the air parcels stayed over Northern Europe for some time after a very fast flow over the North Sea, which ~~corresponds to peaks in~~agrees with the spikes found in the surface SO_2 records observed over the Netherlands during a period of several days.

The high SO_2 concentrations, which ~~that~~ were recorded almost simultaneously at stations over Europe in various sites during the period 21-29 September 2014, are ~~therefore~~ thus associated with the activity of Bárðarbunga volcano (Ialongo et. al., 2015; Table A1 see Appendix A). This is also supported by the back trajectories analysis performed with the HYSPLIT dispersion model that is shown in Figure ~~4~~3. All back trajectories start at 12:00 UTC on the day of maximum SO_2 observations for each one of the Brewer stations and indicate that the arrival of air masses originated from Iceland.

As shown in Figure 4a, the SO_2 plume was detected by the Brewer instruments located in the passage of the volcanic SO_2 plume and from different ground based networks. However, no co-incident measurements were available from the OMI and GOME-2 overpasses at the time of the high SO_2 excursions. Also it should be noted here that no enhanced SO_2 columns were detected by the Brewers located outside of the geographical area covered by the volcanic plume (Fig. 4b). In all volcanic cases we have applied a criterion according to which each daily average from either OMI or GOME-2 should be calculated if and only if more than half of the individual overpasses had data at a given day.~~As shown in Figure 5, the SO_2 plume was detected by instruments under the plume from different ground based networks, e.g. the Brewer instruments, and from OMI and GOME-2 overpasses which were not so clear in this case. The eruption took place at the beginning of September 2014 and several European countries experienced high concentrations of SO_2 at ground level during September. Figure 6 shows similarly the response of ground-level air-base stations under the plume located within 150 km from the nearest Brewer station together with the Brewer measurements.~~

The eruption took place at the beginning of September 2014 and several European countries experienced high concentrations of SO_2 at ground level during the rest of September. Figure 5 shows the response of ground-level AirBase stations under the plume located within 150 km from the nearest Brewer station plotted together with the co-incident Brewer SO_2 column measurements. Interestingly, it suggests that the highest~~Interestingly, it appears that the high~~ amount of SO_2 column measured by the majority of the Brewers during 21 September 2014 due to the volcano reached the surface with a time lag of about one day. The high volcanic concentrations were successfully measured by the ground-based Airbase network. Due to strong European efforts over the last decades to reduce SO_2 emissions, high concentrations of SO_2 are now quite rare in Western Europe (e.g. Vestreng et al., 2007) except in specific areas affected by industrial or shipping emissions. In-situ air quality stations observed high values of SO_2 at the ground level~~,~~ in the coast of France, in the United Kingdom, the Netherlands and Germany between 21 and 25 September 2014. This all ~~pointed~~ points towards ~~an~~ a volcanic episode with a large spatial extent.

~~The high SO_2 columnar concentrations observed at a number of Brewer stations under the volcanic SO_2 plume are shown in Figures 5(a)-(c), averaged from 21 Brewer stations (14 under and 7 outside of the plume) in Europe in Dobson Units (DU). For comparison, SO_2 total columns from OMI (TRM) and GOME-2 daily averages are also plotted on the same figures. For the case of Bárðarbunga, where the volcanic SO_2 was transported in the lowermost troposphere (Schmidt et al., 2015), we also present OMI PBL data, as they were found to agree better with Brewer retrievals than other OMI products e.g. in Sodankylä (Ialongo et al., 2015).~~

As can be seen from Figure 4a, the highest SO_2 column departures from the pre-volcanic baseline were observed from 21 to 22 September 2014. The mean SO_2 column measured by the Brewers under the plume was 2.4 ± 0.8

DU, which was five times greater than the mean column of SO_2 measured by the Brewers outside of the plume (-0.1 ± 0.1 DU) by 2.5 DU on average. The "error bars" show the standard deviation of the daily SO_2 values of all stations during the non-perturbed 10 day period prior to the volcanic eruption. These differences provide rough estimates of the additional SO_2 loading induced by the volcanic eruption over Europe which exceeds 3σ. Comparison between satellite data and Brewer are limited for interpretation because satellite measurements are sparse, represent an average SO_2 column over a relatively large satellite pixel, while the Brewer observations are designed to provide a local point measurement. In spite of the sparsity of OMI observations post Bárðarbunga volcanic eruption, satellite data were used for assimilation in the SO_2 analyses and forecasts produced with the MACC (Monitoring Atmospheric Composition and Climate) system (http://atmosphere.copernicus.eu/). This near-real-time forecasting system assimilates satellite observations to constrain modelling forecasts (Inness et al., 2015; Flemming et al., 2015). The OMI instrument aboard the AURA satellite provided information about concentrations of volcanic SO_2 emitted by the Icelandic Bárðarbunga volcano on 20 September; these observations were assimilated in 2014 by the MACC system in cases of volcanic eruptions, i.e. when OMI values exceeded 5 DU. As shown in Figure 6 (the charts of total column SO_2 are taken from the website http://atmosphere.copernicus.eu/) the subsequent forecasts capture the transport of the plume of volcanic SO_2 southward, while spreading over the continent on 21 and 22 September. The plume stretched all the way from Finland through Poland, Germany and France, to southern England. A parallel forecast, for which no OMI data were used (Fig. 6, right), did not show any elevated SO_2 values, confirming that 'normal' emissions of SO_2 (including shipping and industrial activities) could not explain the observed situation.~~As can be seen from Figure 5, the highest SO_2 columns were observed from 21 to 22 September 2014. The mean SO_2 column measured by the Brewers under the plume was 3.0 ± 0.8 DU, which was greater than the mean column of SO_2 measured by the Brewers outside of the plume (0.6 ± 0.2 DU) by 2.4 DU on average. The "error bars" show the standard errors of the daily values of the stations. The estimates from OMI (PBL) were as follows: mean SO_2 under the plume (0.3 ± 0.5 DU), mean SO_2 outside of the plume (-0.6 ± 0.5 DU), the difference of which is 0.9 DU on average. These differences provide rough estimates of the additional SO_2 loading induced by the volcanic eruption over Europe. The respective estimates from GOME-2 for the period 21-22 September are as follows: mean SO_2 under the plume (0.5 ± 0.2 DU) and mean SO_2 outside of the plume (0.2 ± 0.1 DU). The estimates from OMI (TRM) were accordingly, mean SO_2 under the plume -0.2 ± 0.1 DU, mean SO_2 outside of the plume -0.1 ± 0.1 DU. We note here that the estimates from OMI and GOME-2 are smaller than the estimates from the Brewers. Differences can be attributed to the different measuring techniques and air-mass factors of the SO_2 column and can be caused by uncertainties in both satellite and Brewer measurements. Also, the satellite measurements refer to an average SO_2 column over a relatively large satellite pixel while the Brewer observations refer to local point measurements. In all cases however, the observed SO_2 columns at the stations under the plume were always higher than the columns outside of the plume, which provides important clues as to our capability to detect SO_2 plumes of volcanic origin from ground and space based measurements and also to study them by way of model calculations.~~

~~The above findings were also confirmed by SO_2 analyses and forecasts produced with the MACC (Monitoring Atmospheric Composition and Climate) system (http://atmosphere.copernicus.eu/). This near-to-real-time forecasting system assimilates satellite observations to constrain modelling forecasts (Inness et al., 2015; Flemming et al., 2015). The OMI instrument on board the AURA satellite provided information about~~

~~concentrations of volcanic SO_2 emitted by the Icelandic Bárðarbunga volcano on 20 September; these observations were assimilated in 2014 by the MACC system in cases of volcanic eruptions, i.e. when OMI values exceeded 5 DU. As shown by the chart of total column SO_2 obtained from http://atmosphere.copernicus.eu/ (Figure 7), the subsequent forecasts then captured the transport of this plume of volcanic SO_2 southward spreading over the continent on 21 and 22 September. The plume stretched all the way from Finland through Poland, Germany and France, to southern England. A parallel forecast, for which no OMI data were used, did not show any elevated SO_2 values, confirming that 'normal' emissions of SO_2 (including shipping and industrial activities) could not explain the observed situation.~~

Finally, it should be mentioned here that the thin aerosol layer that has been detected by the PollyXT lidar (Engelmann et al., 2015) over Leipzig at around 2-3 km on 23 and 24 of September 2014 was mostly associated with volcanic ash advection (Figure ~~8~~7). A corresponding cluster analysis of all 155 hourly HYSPLIT back trajectories during this period and for the heights of the layer detected by the lidar (~2.5-3.5km) is shown in Figure ~~9~~8. The increased wind shear that is evident between these heights does not allow a robust characterization of the air masses. However, the source contribution of about 20% from Icelandic air masses supports the volcanic origin of the detected plume. During volcanic eruptions, ash and SO_2 may ~~end up at~~be injected to different altitudes and may follow different trajectories for long-range transport. EARLINET lidars can provide alerts on volcanic ash dispersion over Europe, especially when the systems are employed with depolarization capabilities (e.g. Pappalardo et al., 2013). For the Brewer network capabilities and the Hohenpeissenberg station, Figures ~~8~~ 7 and ~~9~~ 8 demonstrate that ~~the same~~similar approach can be applied ~~which could~~to contribute towards an early warning synergistic tool, as evidenced in the ~~example of the~~ Bárðarbunga case study. The role of the Brewer stations in this system will be the early detection of SO_2 plumes ~~as long as they arrive~~transported over continental areas ~~and the triggering~~that would trigger ~~of~~ the associated forecasting systems (models and networks).

3.2 The 2011 Nabro Volcano plume

A major eruption of Mt. Nabro, a 2218 m high volcano on the border between Eritrea and Ethiopia (13.37^{o} N, 41.7^{o} E~~13.37 °N, 41.7 °E~~), occurred on 12–13 June, 2011. The volcanic eruption injected ash, water vapour and an estimated 1.3–2.0 Tg of SO_2 into the upper troposphere and lower stratosphere (Fairlie et al., 2014 and references therein). In the first phase of the eruption, the main transport pattern of emitted SO_2 followed the strong anticyclonic circulation over the Middle East and Asia associated with the Asian summer monsoon at that time of year (Clarisse et al., 2014 and references therein). In the first month after the eruption stratospheric aerosols were mainly observed over Asia and the Middle East, and by day 60 they covered the whole Northern Hemisphere. ~~Reported~~ Estimated aerosol altitudes from various instruments were between 12 and 21 km (Clarisse et al., 2014). By July 2011 Nabro had cumulatively emitted 5 to 10 percent of what was released by Mount Pinatubo in 1991 (~20 Tg) ranking it among the largest SO_2 emissions in the tropical stratosphere (up to at least 19 km) since Pinatubo (Krotkov et al., 2011). Sulfur dioxide~~SO_2~~ signals of volcanic origin were detected both by Brewer and satellite measurements over ~~eastern~~ East Asia where the volcanic SO_2 plume was transported, as ~~can be seen from~~demonstrated in Figure ~~10~~ 9 and Figure ~~11a~~10a. Measurements ~~come from~~were taken by Brewer in Taipei, Taiwan, ~~in~~ Asia. This is also evident from the back trajectories analysis performed with the HYSPLIT dispersion model for Taipei (Taiwan) as shown in Figure ~~11a~~10a. The analysis indicates that the upper tropospheric air masses arriving at Taipei on June 19, when the peak in SO_2 is observed, originate from Africa.

The Nabro volcanic plume was mainly transported to the East Asia and was detected by various satellite instruments which provide better spatial coverage than the Brewers. A special case study focuses on discrepancies found between ground based and satellite observations of the volcanic SO_2 plume. Brewer located in Tenerife, Spain detected an increase in the SO_2 column, which was not clearly detected by the OMI and GOME-2 satellite overpasses.~~Although the Nabro volcanic plume was mainly transported to the east into Asia and was detected by various satellite instruments which provide better spatial coverage than the Brewers, we present here an interesting case where an increase in the SO2 column due to the volcanic SO2 plume was not clearly detected by the OMI and GOME-2 satellite overpasses but it was clearly detected by the Brewer instrument in Tenerife.~~

More specifically, Figure ~~11b~~ 10b shows back trajectories from Izaña (Tenerife) during 19-29 June 2011 at 15, 17.5 and 20 km heights. It appears that the upper tropospheric-lower stratospheric air masses arriving at Tenerife during 19-29 June originated from Nabro. In June 2011 the Nabro volcano ash plume was detected by the Micropulse Lidar (MPL) located in Santa Cruz de Tenerife (The Canary Islands, Spain). The volcanic plume height ranged from 12 km on 19 June ~~19th~~ to 21 km on 29 June ~~29th~~ (Sawamura et al., 2012). The daily mean SO_2 record (Figure ~~12~~11) shows a 0.5 DU increase at the beginning of the event (19 June ~~19th~~), reaching 0.75 DU on 29 June ~~29th~~ when the layer is found at higher altitude. The signal is not strong and is near the error of 0.5 DU estimated for SO_2 measurement (Stanek, personal communication) but the observations are consistent (independent of the ozone and air mass), since we perform about 100 O_3/SO_2 measurements/day obtaining reduced standard errors associated with daily means as compared to individual observations. The Langley calibration is tracked between calibrations by measurements of the internal lamp (Langley and lamp are shown in Supplement Figure S1). The increase in SO_2 due to the passage of the Nabro volcano plume over the Canary Islands is significant using both methods, showing an offset between them (Figure 12). It is clearly shown that the zero-calibrated Brewer SO_2 data do not compare well with OMI and GOME-2 levels. Instead, the Langley calibrated Brewer data compare better with OMI and GOME-2 retrievals.

In this case the Brewer at Izaña has been able to detect an SO_2 plume at high altitude from a volcano located 7,000 km from the Canary Islands, indicating that the Brewer network is sensitive enough to be incorporated in columnar SO_2 monitoring from volcanic eruptions in worldwide networks.

The case of the 2011 Nabro eruption shows an example of the importance of the Brewer spectrophotometers in measuring and detecting changes in SO_2 amounts in the atmosphere due to volcanic eruptions, in cases where signal in the satellite overpasses is low~~there is poor signal by the satellite overpasses~~. This is true for the case of Izaña (Tenerife) where it appears that OMI and GOME-2 did not clearly detect increases in SO_2 column of volcanic origin between 19 June and 1 July~~19/06 and 01/07~~ as it was the case with the Brewer instrument (Figure ~~12~~11). During some days between 19 June and 1 July~~19/06 and 01/07~~, the Brewer SO_2 columns at Izaña rose above the uncertainty of 0.5 DU for the Brewer SO_2 measurements at ~~Izaña~~this station, whereas the satellite SO_2

columns stayed mostly within the uncertainty of 0.4 DU estimated for OMI (TRM) and GOME-2 satellite retrievals.

These findings can provide clues on the detection limits of such events from a well calibrated Brewer network and a space-borne~~space born~~ instrument. They need further clarification with more Brewers and a larger number of cases.

~~**3.3 The 2011 Grímsvötn volcano case**~~

~~The Grímsvötn volcano (64.42°N, 17.33°W, 1725 m asl) is one of the most active and well-known volcanoes on Iceland. Over the past century, Grímsvötn has erupted about once per decade, the last major eruptions occurring in 1934, 1983, 1996, 2003 and 2011 (http://www.volcano.si.edu) (Moxnes et al., 2014). Note that the Grímsvötn 2011 volcanic eruption is an interesting example of a clear separation of~~ ~~SO_2~~ ~~(transporting mostly northwestward) and the fine ash (transported mostly southeastward) (Moxnes et al., 2014). As expected from the work by Moxnes et al. (2014) we can see that none of the European Brewer stations operating during and after the Grímsvötn eruption were under the volcanic~~ ~~SO_2~~ ~~plume (forward trajectories from Iceland do not pass over the Brewers as can be seen in Figure 13). The average~~ ~~SO_2~~ ~~columnar measurements from 17 Brewer stations in Europe are shown in Figure 14. One can see from both trajectories and measurements that there was no effect in columnar~~ ~~SO_2~~ ~~from that volcanic eruption over Europe.~~

~~3.4~~3.3 The case of the 2010 Eyjafjallajökull volcanic eruption

The Eyjafjallajökull volcano, Iceland (63.63° N, 19.6215° W~~63.63°N, 19.6215°W~~; 1666 m a.s.l.) erupted explosively on 14 April 2010 and continued to emit ash and gas until 24 May (Flentje et al., 2010; Thomas and Prata, 2011; Stohl et al., 2011; Flemming and Inness, 2013). Despite the relatively modest size of the eruption, the prevailing wind conditions advected the volcanic plume to the south-east leading to unprecedented disruption to air traffic in Western Europe. This caused significant financial losses for the airlines and highlights the importance of efficient volcanic cloud monitoring and forecasting. Results demonstrate that the eruption can be divided into an initial ash rich phase (14-18 April), a lower intensity middle phase (19 April until early May) and a final phase (4-24 May) where considerably great quantities both ash and SO_2 were released (Thomas and Prata, 2011).

Figure ~~15~~ 12 shows the responses of Brewer stations under the volcanic SO_2 plume and the average of Brewer stations outside of the plume together with OMI and GOME-2 satellite observations. We determined 9 stations being under the plume in 2010 and ~~10~~ 11 stations being outside of the plume based on analysis of forward and backward trajectories of air masses following the volcanic eruption. The stations determined to be under the plume in 2010 (shown in Figure 12a) are Sodankÿla, Obninsk, Manchester, De Bilt, Belsk, Reading, Hohenpeissenberg, Davos and Arosa. The stations determined to be outside of the plume are Vindeln, Oslo, Norrkoeping, Copenhagen, Uccle, Hradec Kralove, Aosta, Kislovodsk, Rome, Thessaloniki and Athens (Figure 12b).~~The stations determined to be under the plume in 2010 (shown in Figure 15b) were Belsk, De Bilt, Hohenpeissenberg, Obninsk, Sodankÿla, Davos, Manchester, Reading and Arosa. The stations determined to be outside of the plume were Athens, Aosta, Copenhagen, Hradec Kralove, Kislovodsk, Thessaloniki, Uccle,~~

~~Norrkoeping and Vindeln (Figure 15c).~~ It may seem ~~a bit~~ surprising that Uccle and De Bilt fall in different categories as they are ~~close together~~closely located, but the data did not show increased SO_2 at Uccle during days when increased SO_2 was measured at De Bilt. In Table A1 of Appendix A, we present the dates ~~in which~~when the examined Brewer stations were ~~determined to be~~ either under or outside of the volcanic SO_2 plume. ~~according to careful~~Careful analysis of the trajectories of the volcanic plumes in 2010 and 2014 helped verify these analyses. The distinction between stations outside of the plume and stations under the plume was done as follows: whenever SO_2 at each station measuring exceeded 2 DU (2σ) back trajectories were calculated and the origin was compared to the location of the volcanic eruption.~~At each station measuring SO_2 exceeding 2 DU (2σ) we calculated back trajectories and found that their origin was at the volcanic eruption.~~ All these stations have been considered to be under the SO_2 plume. All other stations, for which columnar SO_2 amounts were within 2σ and were not originating from the area of the eruption, were considered to be outside of the volcanic SO_2 plume.

As we can see from Figure ~~15~~12, the columnar SO_2 departures~~average SO_2 amount~~ at stations located under the passage of the volcanic SO_2 plume exceeded 0.3 DU (reaching 1.5 DU in some cases) whereas at stations located outside of the plume, the columnar SO_2 departures~~columns~~ did not exceed 0.3 DU~~ on average~~. Moreover, during the explosive phase 2 there were three main periods in which the volcanic aerosol content was observed by EARLINET over Europe: 15-26 April, 5-13 May and 17-20 May. These periods were determined from measurements of the integrated backscatter at 532 nm in the volcanic layers (Pappalardo et al., 2013). We estimate high SO_2 columnar departures~~that the average SO_2 columns~~ measured by the Brewers under the plume during these three periods up to 6.0 DU (e.g. Arosa, 18 May 2016).~~were 0.3 ± 0.1 DU, 0.2 ± 0.2 DU and 0.8 ± 0.3 DU, respectively.~~

We note here that the ash cloud caused further disruptions to air transportation on 4-5 May and 16-17 May 2010, particularly over Ireland and the UK. The average SO_2 ~~columns~~ columnar departures measured by the Brewers under the plume in the UK (Manchester and Reading) during these two periods were estimated to ~~0.1~~1.1 ± ~~0.4~~0.3 DU and ~~1.0~~1.5 ± ~~0.5~~0.4 DU respectively~~, both within the error bars~~. These amounts were higher than the amounts measured outside of the plume (~~-0.2~~-0.1 ± ~~0.3~~0.2 DU and ~~0.2~~-0.1 ± ~~0.2~~0.1 DU, accordingly) almost by ~~0.5~~1.4 DU on average.

~~3.5~~3.4 An eruption of larger scale importance – The 2008 Kasatochi case

The eruption of Kasatochi volcano on 7-8 August 2008 injected large amounts of material and SO_2 into the troposphere and lower stratosphere of the northern middle latitudes during a period of low stratospheric aerosol background concentrations. The Kasatochi volcano in the central Aleutian Islands of Alaska (52.17° N, 175.51° W~~52.17°N, 175.51°W~~) erupted three times between 2201 UTC on 7 August and 0435 UTC on 8 August 2008 (Bitar et al., 2010). Aerosols from the volcanic eruption were detected by lidar in Halifax shortly after the eruption (Bitar et al., 2010). The total mass of SO_2 injected into the atmosphere by the eruption is estimated to 1.7 Tg, with about 1 Tg reaching the stratosphere (above 10 km asl) (Kristiansen et al., 2010).

We have studied the columnar SO_2 amounts following the Kasatochi eruption in August 2008 from ground based and satellite data. Figure 13 shows the columnar SO_2 departures from the unperturbed 10 day pre-volcanic period over Canada/USA and Europe during the bimonthly period August-September 2008 as measured by the Brewers in comparison with the satellite observations by OMI and GOME-2.~~Figure 16 shows the columnar SO_2 amounts over Canada/USA, Europe and Taiwan during the bimonthly period August-September 2008 in two panels. Figure 16a shows the SO_2 columns as measured by the Brewers (together with a 7-day running mean which was applied to the data), and Figure 16b shows the Brewer measurements in comparison with the satellite observations by OMI and GOME-2.~~

The SO_2 plume was clearly seen by the Brewers in Canada/USA (Figure ~~16a~~13) and it was also detected by the majority of the Brewers in Europe with a delay ~~of~~ by about 3 days. The total SO_2 columnar departures averaged over Canada during the period 12-20 August 2008 ~~is~~ are estimated to ~~0.8~~0.9 ± 0.3 DU~~, which is 1.1 DU more than the background atmospheric SO_2 column in Canada (-0.3 ± 0.1 DU)~~. Accordingly over Europe, we estimate a mean SO_2 columnar departure of ~~1.4~~1.0 ± 0.1 DU during the period 15-22 August 2008~~ and a background mean of 0.4 ± 0.02 DU~~. ~~Their difference of 1.0 DU~~This number gives a rough estimate of the average volcanic SO_2 column measured by the Brewers over Europe. We note here that the e-folding time of the Kasatochi SO_2, i.e. the time where the volcanic SO_2 amount decayed, was estimated to be about 8-9 days (Krotkov et al., 2010).~~We note here that the 7-day running mean filter was applied to the data as a better visualization of the periods with increased SO_2 concentrations in the atmosphere after the Kasatochi eruption. The curve roughly coincides with the e-folding time of the SO_2 column i.e. the time where the volcanic SO_2 amount decayed. Indeed, the e-folding time of the Kasatochi SO_2 was estimated to be about 8-9 days (Krotkov et al., 2010).~~

The high amounts of SO_2 and the variability of SO_2 measured in Europe by the Brewers after the eruption of Kasatochi in August 2008 are in line with OMI (TRM) and GOME-2 satellite observations. More specifically, OMI (TRM) shows an average SO_2 columnar departure of 0.5 ± 0.1 DU during the period 15-22 August 2008 and GOME-2 an average SO_2 columnar departure of 0.8 ± 0.1 DU respectively.~~More specifically, OMI (TRM) shows an average SO_2 column of 0.5 ± 0.1 DU during the period 15-22 August 2008 and a background mean of -0.02 ± 0.01 DU. The respective values from GOME-2 are 0.8 ± 0.1 DU for the volcanic period 15-22 August and 0.2 ± 0.01 DU for the background atmosphere.~~

The Brewer data have been correlated with that from OMI and GOME-2. The Pearson's correlation coefficients between the three datasets were all highly statistically significant (>99%). The correlation between SO_2 from the Brewers and SO_2 from GOME-2 at 19 stations averaged over Europe is +0.86 (t-value = 12.54, p-value < 0.0001, N = 59).~~+0.933 (t-value = 13.98235, p-value < 0.0001, N = 31).~~ Accordingly, the correlation between Brewer and OMI (TRM) SO_2 data is +0.86 (t-value = 11.77, p < 0.0001, N = 50)~~+0.919 (t-value = 12.34644, p < 0.0001, N = 30)~~ and between GOME-2 and OMI (TRM) data is +0.92 (t-value = 16.32, p < 0.0001, N = 48).~~+0.922 (t-value = 12.63061, p < 0.0001, N = 30).~~ These correlations were calculated from ~~30~~ 60 daily averages during the Kasatochi volcanic eruption in August-September 2008. The statistical tests gave significant results and verified the capability of the Brewers in detecting natural SO_2 emitted by volcanoes when the volcanic plume of SO_2 passes over the ground sites. We note here that there is a general consistency between ~~the~~ all three datasets (Brewers, OMI and GOME-2) on the changes in SO_2 following the Kasatochi volcanic eruption~~, Brewers, the OMI and GOME-2 estimates~~.

Table 5 summarises the correlation coefficients between the mean columnar SO_2 measured by all Brewers ~~in the Northern Hemisphere~~over Europe and provided by the satellite products of OMI and GOME-2 during the globally ~~important~~ extended Kasatochi event. The correlation coefficients have high statistical significance explaining more than ~~80%~~70% of the total variance between the columnar SO_2 measurements from ground and space in the case of Kasatochi. However, the discrepancies found between satellite and Brewer observations during the other volcanic eruptions could be impacted by sparsity of coincident measurements, and thus cannot confirm or deny Kasatochi case findings at high significance levels.

4 Conclusions

In this work we provide evidence that the current network of Brewer spectroradiometers is capable of identifying columnar SO_2 plumes of volcanic origin. The study is based on the results from the three largest volcanic eruptions (VEI ≥4) in the past decade when elevated SO_2 plumes have passed over Brewer stations in the Northern Hemisphere. The analysis included a fourth eruption, namely Bárðarbunga, because it has perturbed the SO_2 regime over large parts of Europe and extended from the free troposphere down to the surface. Back and forward trajectory analysis have been used to aid in identifying and selecting measurements taken under and outside of the volcanic SO_2 plume. When the plume was overpassing a site, the SO_2 signal was found to be quite high, exceeding 3σ of daily values relative to the average levels taken during the unperturbed measurements over ten days preceding each eruption. On the average the mean SO_2 columnar amount to be attributed to the volcano is estimated to be on the order of 2 DU as discussed in section 3. In addition to the Brewer network, comparisons were made with other instruments (e.g. surface SO_2 sensors) that were located under the volcanic SO_2 plumes. Moreover, satellite measurements of columnar SO_2 from OMI and GOME-2 collocated with the Brewer network were used for comparisons.

From the results discussed in section 3 some general remarks can be put forward concerning SO_2 levels and detection time after the eruption. Starting with the Kasatochi eruption, as it appears from Figure 13, the plume can be detected 4 days after the eruption over Canada and the US and about 7 days over Europe with an average amplitude on the order of 2 DU compared to the unperturbed ten day pre-volcanic period (baseline). All estimates are based obviously on measurements taken under the plume. The Kasatochi eruption provided a formidable example for a volcanic SO_2 plume to be observed not only by the ground based instruments, but from space-borne as well (OMI and GOME-2). Relative to the undisturbed period before Kasatochi the amplitude of the signal is 2 DU for GOME-2 and 1.5 DU for OMI. The results for the other volcanic eruptions are similar for the Brewer network, but unfortunately because of the sparsity of satellite overpassing the Brewer stations the satellite data concur with those from the Brewers only in Kasatochi. Based on the above discussion it appears that currently no single network can independently and fully monitor the evolution of volcanic SO_2 plumes. Among a few reasons are lack of measurements during peak values, complications from meteorological events, ejection heights and exposure conditions. The evidence presented here points that combination of observations from various instruments, aided by chemical transport models and operated in synergy could address such a complex issue.

~~In this work we provide strong evidence that the current network of Brewer spectroradiometers is capable of identifying columnar SO_2 emissions of volcanic origin. The study was based on the results from the five largest volcanic eruptions in the past decade and the analyses was confined to the Northern Hemisphere where the Brewer network is more dense. The sensitivity of that network to detect volcanic SO_2 plumes was shown to be quite different depending on the strength and the trajectory of the plume. If the plume is overpassing the site, the signal to noise ratio was found to be quite high, exceeding 2σ or more of the daily means. In addition, volcanic eruptions of regional importance could be observed in detail down to ground level (e.g. Bárðarbunga). The statistical findings with the Brewer network have been compared to independent measurements by satellites and our conclusions also rely on information gathered through modeling tools. The comparison with satellite measurements shows statistically tested agreement between the Brewer network and collocated measurements of columnar SO_2 from OMI and GOME-2. Moreover, additional aid was provided by other independent networks such as the EARLINET and the AirBase.~~

The combination of the above discussed observation and modelling tools can assist in detecting existing volcanic plumes, but also in forecasting their evolution, which can have importance not only to the air traffic warning but also to air pollution in the lower layers of the atmosphere.~~In synergy all of these tools, are capable not only to detect existing volcanic plumes but also to forecast their evolution, can have importance not only to air traffic but also to air pollution in the lower layers of the atmosphere.~~ Therefore, an automated source receptor modelling tool could be proposed as follows: a modelling system based on FLEXPART and HYSPLIT backward-trajectory simulations could be automatically triggered whenever high SO_2 values are detected at a Brewer station above a specific threshold (e.g. ~~2σ~~3σ of station's daily values) or when a lidar instrument detects highly depolarizing layers that were not advected from a geographical location over a desert. The operational use of such a synergistic activity could provide ~~near-to-real time~~near-real-time and forecasting information on the evolution of volcanic episodes and also develop a comprehensive database of measurements useful to improve model ~~results~~forecasts. This new well-tuned and ~~organized~~ organised synergistic activity of monitoring networks, observations and modelling from ground and space could create a challenging monitoring tool for volcanic and other extreme emissions, which form the basis towards a new regional SO_2 forecasting ~~tool~~facility.

5 Data availability

SO_2 columns at Churchill, Goose, Edmonton, Regina, Saturna Island and~~,~~ Toronto in Canada, ~~and~~ Taipei in Taiwan, Boulder and Mauna Loa in the US were obtained from the World Ozone and Ultraviolet Radiation Data Centre (WOUDC; http://www.woudc.org/, last access: 10 October 2016)~~. SO_2 columns at Niwot Ridge, USA, were downloaded from~~ and the NOAA-EPA Brewer Spectrophotometer UV and Ozone Network (NEUBrew; http://www.esrl.noaa.gov/gmd/grad/neubrew/, last access: 10 October 2016). OMI and GOME-2 satellite SO_2 data products were downloaded from the Aura Validation Data Center (AVDC) ~~at the website~~ (available from: http://avdc.gsfc.nasa.gov/index.php?site=245276100, last access: 10 October 2016). Surface SO_2 concentrations over Europe were acquired from the European Environment Agency databases (AirBase) (http://www.eea.europa.eu/data-and-maps/data/aqereporting-1#tab-european-data, last access: 10 October 2016).

Appendix A

Table A1. Dates ~~where~~ in which the Brewers were determined to be under or outside of the volcanic SO_2 plume, based on analysis of back trajectories of the volcanic plumes in 2010 and 2014. The distinction between stations outside of the plume and stations under the plume was done as follows: At each station measuring SO_2 exceeding 2 DU (2σ) we calculated back trajectories and found that their origin was at the volcanic eruption. All these stations have been considered to be under the SO_2 plume. All other stations, for which columnar SO_2 amounts were within 2σ and were not originating from the area of the eruption, were considered to be outside of the volcanic SO_2 plume. During the Kasatochi eruption all Brewers ~~are~~ were considered to be under the volcanic SO_2 plume ~~while during Grímsvötn eruption all Brewers are considered to be outside of the plume~~.

Station	LAT (deg)	LON (deg)	ALT (m)	2010	2014
Sodankylä	67.36	26.63	180	20/4	27/9 and 29/9
Vindeln	64.24	19.77	225	Outside the plume	29/9
Jokioinen	60.82	23.50	106	No data	27/9
Oslo	59.90	10.73	50	Outside the plume	~~27/9~~Outside the plume
Norrkoeping	58.58	16.15	43	Outside the plume	30/9
Copenhagen	55.63	12.67	50	Outside the plume	~~26~~24/9
Obninsk	55.10	36.60	100	23/4 and 25/4	28/9
Manchester	53.47	-2.23	76	16/5	21/9
Warsaw	52.17	20.97	107	No data	Outside the plume
De Bilt	52.10	5.18	2	2/5, 11/5, 18/5	21/9
Belsk	51.84	20.79	180	10/5	Outside the plume
Reading	51.44	-0.94	66	16/5	21/9
Uccle	50.80	4.36	100	Outside the plume	21-22/9
Hradec Kralove	50.18	15.84	285	Outside the plume	~~29~~24/9
Hohenpeissenberg	47.80	11.01	985	18/5	22/9
Davos	46.81	9.84	1590	27/4 and 18-19/5	Outside the plume
Arosa	46.78	9.67	1840	18/5	Outside the plume
Aosta	45.74	7.36	569	Outside the plume	~~21/9 and~~ 23/9
Kislovodsk	43.73	42.66	2070	Outside the plume	Outside the plume
Rome	41.90	12.52	75	~~Data not used~~Outside the plume	Outside the plume
Thessaloniki	40.63	22.95	60	Outside the plume	No data
Athens	37.99	23.78	191	Outside the plume	Outside the plume

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Table 1. Volcanic eruptions in the past decade considered in this study.

Volcano	Latitude	Longitude	Elevation (asl)	Period of Eruption	VEI*
Okmok, Alaska	53.43°N	168.13°W	1073 m	12 July - 19 August 2008	4
Kasatochi, Alaska	52.17°N	175.51°W	300 m	7-8 August 2008	4
Sarychev, Russia	48.1°N	153.2°E	1496 m	12-17 June 2009	4
Eyjafjallajökull, Iceland	63.63°N	19.62°W	1666 m	14 April - 23 May 2010	4
Grímsvötn, Iceland	64.42°N	17.33°W	1725 m	21-25 May 2011	4
Nabro, Africa	13.37°N	41.70°E	2218 m	12-13 June 2011	4
Tolbachik, Russia	55.83°N	160.33°E	3611 m	27 November 2012 - 22 August 2013	4
Bárðarbunga, Iceland	64.64°N	17.56°W	2005 m	31 August 2014 - 28 February 2015	0

*taken from the Smithsonian Institution Global Volcanism Program

~~**Table 1. The 5 major volcanic eruptions in the past decade analysed in this study.**~~

~~Volcano~~	~~Latitude~~	~~Longitude~~	~~Elevation (asl)~~	~~Period of Eruption~~
~~Kasatochi, Alaska~~	~~52.17°N~~	~~175.51°W~~	~~300 m~~	~~7-8 August 2008~~
~~Eyjafjallajökull, Iceland~~	~~63.63°N~~	~~19.62°W~~	~~1666 m~~	~~14 April – 23 May 2010~~
~~Grímsvötn, Iceland~~	~~64.42°N~~	~~17.33°W~~	~~1725 m~~	~~21-25 May 2011~~
~~Nabro, Africa~~	~~13.37°N~~	~~41.70°E~~	~~2218 m~~	~~12-13 June 2011~~
~~Bárðarbunga, Iceland~~	~~64.64°N~~	~~17.56°W~~	~~2005 m~~	~~31 August 2014 – 28 February 2015~~

Table 2. Stations with accessible SO_2 column data from Brewers analysed in this study. Stations are sorted from high to lower northern latitudes.

	Station	Latitude	Longitude	Elevation asl (m)	Instruments	Data source
1	Sodankÿla	67.36	26.63	180	Brewer MKII 037	FMI
2	Vindeln	64.24	19.77	225	Brewer MKII 006	SMHI
3	Jokioinen	60.82	23.50	106	Brewer MKIII 107	FMI
4	Oslo	59.90	10.73	50	Brewer MKV 042	U_Oslo
5	Churchill	58.74	-93.82	16	Brewer MKII 026, Brewer MKIV 032, Brewer MKIII 203	WOUDC
6	Norrkoeping	58.58	16.15	43	Brewer MKIII 128	SMHI
7	Copenhagen	55.63	12.67	50	Brewer MKIVe 082	DMI
8	Obninsk	55.10	36.60	100	Brewer MKII 044	IEM-SPA
9	Edmonton	53.55	-114.10	766	Brewer MKII 055, Brewer MKIV 022	WOUDC
10	Manchester	53.47	-2.23	76	Brewer MKIII 172	U_Manchester
11	Goose Bay	53.29	-60.39	39	Brewer MKII 018	WOUDC
12	Warsaw	52.17	20.97	107	Brewer MKIII 207	PAS-IGF
13	De Bilt	52.10	5.18	24	Brewer MKIII 189	KNMI
14	Belsk	51.84	20.79	180	Brewer MKII 064	PAS-IGF
15	Reading	51.44	-0.94	66	Brewer MKIV 075, Brewer MKII 126	U_Manchester
16	Uccle	50.80	4.36	100	Brewer MKII 016, Brewer MKIII 178	RMIB
17	Regina	50.20	-104.71	580	Brewer MKIII 111	WOUDC
18	Hradec Kralove	50.18	15.84	285	Brewer MKIII 184	CHMI-HK
19	Saturna Island	48.78	-123.13	178	Brewer MKII 012	WOUDC
20	Hohenpeissenberg	47.80	11.01	985	Brewer MKII 010	DWD-MOHp
21	Davos	46.81	9.84	1590	Brewer MKIII 163	PMOD/WRC
22	Arosa	46.78	9.67	1840	Brewer MKII 040, Brewer MKIII 156	MeteoSwiss
23	Aosta	45.74	7.36	569	Brewer MKIV 066	ARPA-VDA
24	Toronto	43.78	-79.47	198	Brewer MKII 015	WOUDC
25	Kislovodsk	43.73	42.66	2070	Brewer MKII 043	RAS-IAP
26	Rome	41.90	12.52	75	Brewer MKIV 067	U_Rome
27	Thessaloniki	40.63	22.95	60	Brewer MKII 005	AUTH
28	Boulder	40.03	-105.53	2891	Brewer MKIV 146	NEUBrew
29	Athens	37.99	23.78	191	Brewer MKIV 001	BRFAA
30	Izaña	28.31	-16.50	2373	Brewer MKIII 157	AEMET

31	Taipei	25.04	121.51	5	Brewer MKIII 129	WOUDC
32	Mauna Loa	19.54	-155.60	3397	Brewer MKIII 119	WOUDC

~~**Table 2. Mid-latitude stations with accessible SO_2 column data from Brewers analysed in this study. Stations are sorted from high to lower northern latitudes.**~~

	~~Latitude~~	~~Longitude~~	~~Elevation asl (m)~~	~~Instruments~~	~~Data source~~
~~SODANKYLA~~	~~67.36~~	~~26.63~~	~~180~~	~~Brewer MKII 037~~	~~FMI~~
~~VINDELN~~	~~64.24~~	~~19.77~~	~~225~~	~~Brewer MKII 006~~	~~SMHI~~
~~JOKIOINEN~~	~~60.82~~	~~23.50~~	~~106~~	~~Brewer MKIII 107~~	~~FMI~~
~~OSLO~~	~~59.90~~	~~10.73~~	~~50~~	~~Brewer MKV 042~~	~~U_Oslo~~
~~CHURCHILL~~	~~58.74~~	~~-93.82~~	~~16~~	~~Brewer MKII 026, Brewer MKIV 032, Brewer MKIII 203~~	~~WOUDC~~
~~NORRKOEPING~~	~~58.58~~	~~16.15~~	~~43~~	~~Brewer MKIII 128~~	~~SMHI~~
~~COPENHAGEN~~	~~55.63~~	~~12.67~~	~~50~~	~~Brewer MKIVe 082~~	~~DMI~~
~~OBNINSK~~	~~55.10~~	~~36.60~~	~~100~~	~~Brewer MKII 044~~	~~IEM-SPA~~
~~EDMONTON~~	~~53.55~~	~~-114.10~~	~~766~~	~~Brewer MKII 055, Brewer MKIV 022~~	~~WOUDC~~
~~MANCHESTER~~	~~53.47~~	~~-2.23~~	~~76~~	~~Brewer MKIII 172~~	~~U_Manchester~~
~~WARSAW~~	~~52.17~~	~~20.97~~	~~107~~	~~Brewer MKIII 207~~	~~PAS-IGF~~
~~DE BILT~~	~~52.10~~	~~5.18~~	~~2~~	~~Brewer MKIII 189~~	~~KNMI~~
~~BELSK~~	~~51.84~~	~~20.79~~	~~180~~	~~Brewer MKII 064~~	~~PAS-IGF~~
~~READING~~	~~51.44~~	~~-0.94~~	~~66~~	~~Brewer MKIV 075, Brewer MKII 126~~	~~U_Manchester~~
~~UCCLE~~	~~50.80~~	~~4.36~~	~~100~~	~~Brewer MKII 016, Brewer MKIII 178~~	~~RMIB~~
~~HRADEC KRALOVE~~	~~50.18~~	~~15.84~~	~~285~~	~~Brewer MKIII 184~~	~~CHMI-HK~~
~~SATURNA ISLAND~~	~~48.78~~	~~-123.13~~	~~178~~	~~Brewer MKII 012~~	~~WOUDC~~
~~HOHENPEISSENBERG~~	~~47.80~~	~~11.01~~	~~985~~	~~Brewer MKII 010~~	~~DWD-MOHp~~
~~DAVOS~~	~~46.81~~	~~9.84~~	~~1590~~	~~Brewer MKIII 163~~	~~PMOD/WRC~~
~~AROSA~~	~~46.78~~	~~9.67~~	~~1840~~	~~Brewer MKII 040, Brewer MKIII 156~~	~~MeteoSwiss~~
~~AOSTA~~	~~45.74~~	~~7.36~~	~~569~~	~~Brewer MKIV 066~~	~~ARPA-VDA~~
~~TORONTO~~	~~43.78~~	~~-79.47~~	~~198~~	~~Brewer MKII 015~~	~~WOUDC~~
~~KISLOVODSK~~	~~43.73~~	~~42.66~~	~~2070~~	~~Brewer MKII 043~~	~~RAS-IAP~~
~~ROME~~	~~41.90~~	~~12.52~~	~~75~~	~~Brewer MKIV 067~~	~~U_Rome~~
~~THESSALONIKI~~	~~40.63~~	~~22.95~~	~~60~~	~~Brewer MKII 005~~	~~AUTH~~
~~NIWOT RIDGE~~	~~40.03~~	~~-105.53~~	~~2891~~	~~Brewer MKIV 146~~	~~NEUBrew~~
~~ATHENS~~	~~37.99~~	~~23.78~~	~~191~~	~~Brewer MKIV 001~~	~~BRFAA~~

Table 3. Rural AirBase stations analysed in this study (see text).

Station ID	Station name	Latitude	Longitude	Closest Brewer (within 150 km)
GB0583A	Middlesbrouth	54.569	-1.221	Manchester
NL00444	De Zilk-Vogelaarsdreef	52.298	4.51	Uccle
PL0105A	Parzniewice	51.291	19.517	Belsk
NL00133	Wijnandsrade-Opfergeltstraat	50.903	5.882	De Bilt
GB0038R	Lullington Heath	50.794	0.181	Reading
CH0005R	Rigi	47.067	8.463	Arosa
CH0002R	Payerne	46.813	6.944	Aosta

Table 4. SO_2 column departures at mid-latitude stations averaged in bimonthly periods following volcanic eruptions.

		August-September 2008 (Kasatochi)		April-May 2010 (Eyjafjallajökull)		September-October 2014 (Bárðarbunga)	
(a)	Latitude	mean	σ	mean	σ	mean	σ
Sodankÿla	67.36	0.6	2.1	0.1	0.7	-0.5	1.8
Vindeln	64.24	0.4	1.4	0.0	0.4	-0.2	0.9

Jokioinen	60.82	0.5	0.6	*	*	0.4	0.5
Oslo	59.90	*	*	0.7	0.6	-0.1	1.0
Churchill	58.74	0.6	0.8	-0.3	1.1	0.4	1.0
Norrkoeping	58.58	0.4	0.8	-0.1	0.2	0.1	0.8
Copenhagen	55.63	0.3	0.8	0.5	0.9	-0.4	0.7
Obninsk	55.10	*	*	0.1	0.5	0.3	0.9
Edmonton	53.55	0.4	0.6	0.4	0.4	0.0	0.4
Manchester	53.47	0.6	0.7	0.0	0.6	0.4	1.6
Goose Bay	53.29	0.2	0.4	*	*	0.3	0.3
Warsaw	52.17	*	*	*	*	0.1	0.4
De Bilt	52.10	0.1	0.9	-0.3	0.9	0.2	0.8
Belsk	51.84	0.3	0.6	-0.4	0.4	0.4	0.5
Reading	51.44	0.2	0.7	1.2	1.2	0.3	1.7
Uccle	50.80	0.1	0.6	-0.5	0.6	0.7	1.3
Regina	50.20	0.0	0.9	*	*	*	*
Hradec Kralove	50.18	0.2	0.4	-0.3	0.4	-0.6	0.7
Saturna Island	48.78	0.4	1.1	0.0	0.2	0.4	0.5
Hohenpeissenberg	47.80	0.0	0.5	0.5	0.6	-0.1	1.6
Davos	46.81	0.2	0.5	-0.1	0.3	-0.1	0.2
Arosa	46.78	0.6	1.5	-0.5	1.5	-0.1	0.5
Aosta	45.74	-0.1	0.6	0.0	0.6	-0.6	0.8
Toronto	43.78	0.5	1.0	-0.2	0.5	0.4	0.5
Kislovodsk	43.73	-0.3	0.3	-0.1	0.3	0.2	0.2
Rome	41.90	-0.1	1.1	-0.8	1.3	-0.2	0.5
Thessaloniki	40.63	0.4	0.7	-0.7	0.9	*	*
Boulder	40.03	0.1	0.5	0.1	0.9	*	*
Athens	37.99	0.9	0.8	-0.4	0.6	0.0	0.4
(b)		mean ± st. error (N)		mean ± st. error (N)		mean ± st. error (N)	
All Brewers		0.29 ± 0.03 (1051)		-0.04 ± 0.03 (1064)		0.07 ± 0.03 (861)	
GOME-2		0.23 ± 0.02 (1057)		0.08 ± 0.01 (971)		-0.03 ± 0.02 (677)	
OMI (TRM)		0.15 ± 0.02 (741)		0.00 ± 0.02 (438)		0.01 ± 0.02 (395)	

(*) missing values are those possessing < 25 days of data in each bimonthly period, or no data.

Table 5. Correlation coefficients between the mean columnar SO_2 measured by the brewers in Europe and provided by the satellite products of OMI and GOME-2 during the volcanic eruptions of Kasatochi (2008), Eyjafjallajökull (2011) and Bárðarbunga (2014) for stations located under the volcanic SO_2 plume.

Europe	August-September 2008	April-May 2010	September-October 2014
Brewers and GOME-2	**0.86** [59] (p<0.0001)	**0.31** [54] (p=0.02336)	**0.44** [39] (p=0.00496)
Brewers and OMI (TRM)	**0.86** [50] (p<0.0001)	(*) [16]	(*) [16]
GOME-2 and OMI (TRM)	**0.92** [48] (p<0.0001)	(*) [15]	(*) [15]

Bold: all the above correlations are significant at confidence level 95% or greater (t-test).
(*): missing correlations are those possessing less than 30 days of data in each bimonthly period. In brackets: number of pairs.

Table 4. SO_2 columns at mid-latitude stations averaged in bimonthly periods which include volcanic eruptions.

(a)	Latitude	August-September 2008 mean	σ	N (days)	April-May 2010 mean	σ	N (days)	May-June 2011 mean	σ	N (days)	September-October 2014 mean	σ	N (days)
SODANKYLA	67.36	0.7	1.9	41	-0.5	0.6	44	0.1	0.6	59	0.7	1.8	27
VINDELN	64.24	0.5	1.2	45	0.4	0.4	49	[-3.2]	0.8	56	0.3	0.8	33
JOKIOINEN	60.82	0.4	0.6	42	*	*	*	0.2	0.3	53	0.6	0.5	30
OSLO	59.90	*	*	*	-1.7	0.7	52	0.9	0.8	51	-0.1	0.9	41
CHURCHILL	58.74	0.6	0.9	42	1.5	1.1	47	2.2	0.8	45	0.3	0.9	25
NORRKOEPING	58.58	0.2	0.8	41	0.0	0.2	50	0.7	0.3	59	0.3	0.7	39
COPENHAGEN	55.63	1.6	0.8	55	-0.4	0.9	48	0.7	0.8	31	2.6	0.6	38
OBNINSK	55.10	*	*	*	0.3	0.6	57	0.6	0.4	58	-0.1	0.9	40
EDMONTON	53.55	-0.2	0.5	56	-1.0	0.5	53	1.5	1.1	56	*	*	12
MANCHESTER	53.47	0.6	0.7	35	0.7	0.6	46	0.9	0.5	40	0.1	1.5	31
WARSAW	52.17	*	*	*	*	*	*	*	*	*	0.9	0.4	45
DEBILT	52.10	0.5	0.8	61	0.4	0.9	61	0.0	0.6	61	0.3	0.8	53
BELSK	51.84	1.0	0.5	46	1.1	0.4	45	0.9	0.4	47	0.6	0.5	50
READING	51.44	-0.3	0.7	36	-1.4	1.5	57	1.1	0.6	49	-0.1	1.5	45
UCCLE	50.80	0.7	0.5	46	-0.3	0.6	50	-0.3	0.5	54	1.6	1.2	43
HRADEC KRALOVE	50.18	0.5	0.4	47	0.3	0.4	44	0.4	0.4	52	0.6	0.8	42
SATURNA ISLAND	48.78	-0.4	1.2	53	-0.3	0.2	55	1.4	0.4	54	0.6	0.5	45
HOHENPEISSENBERG	47.80	-0.1	0.5	52	0.4	0.6	48	0.5	0.5	42	0.8	1.4	52
DAVOS	46.81	0.5	0.5	42	0.6	0.3	42	*	*	15	2.0	0.2	55
AROSA	46.78	0.5	1.4	61	1.3	1.8	59	1.7	1.2	61	-0.3	0.6	59
AOSTA	45.74	0.2	0.5	53	0.0	0.6	52	0.2	0.5	29	1.1	0.8	43
TORONTO	43.78	-0.3	0.9	49	-0.6	0.5	52	0.7	1.2	33	1.8	0.5	39
KISLOVODSK	43.73	-0.2	0.3	40	0.3	0.2	49	0.3	0.4	44	0.1	0.2	50
ROME	41.90	1.2	1.0	57	[4.4]	1.2	50	[4.6]	0.6	58	0.5	0.5	56
THESSALONIKI	40.63	0.4	0.7	54	0.9	0.9	49	1.9	1.0	53	*	*	*
NIWOT RIDGE	40.03	-0.4	0.5	56	-1.1	0.9	45	-0.7	0.4	54	*	*	*
ATHENS	37.99	1.6	0.8	55	0.4	0.7	53	[4.3]	1.4	53	0.9	0.4	44
(b)		mean ± st. error			mean ± st. error			mean ± st. error			mean ± st. error		
All Brewers		0.41 ± 0.12			0.05 ± 0.12			0.72 ± 0.15			0.67 ± 0.15		
GOME-2		0.26 ± 0.02			0.01 ± 0.01			0.02 ± 0.01			0.08 ± 0.02		
OMI (TRM)		0.04 ± 0.02			-0.03 ± 0.02			-0.03 ± 0.02			-0.06 ± 0.02		

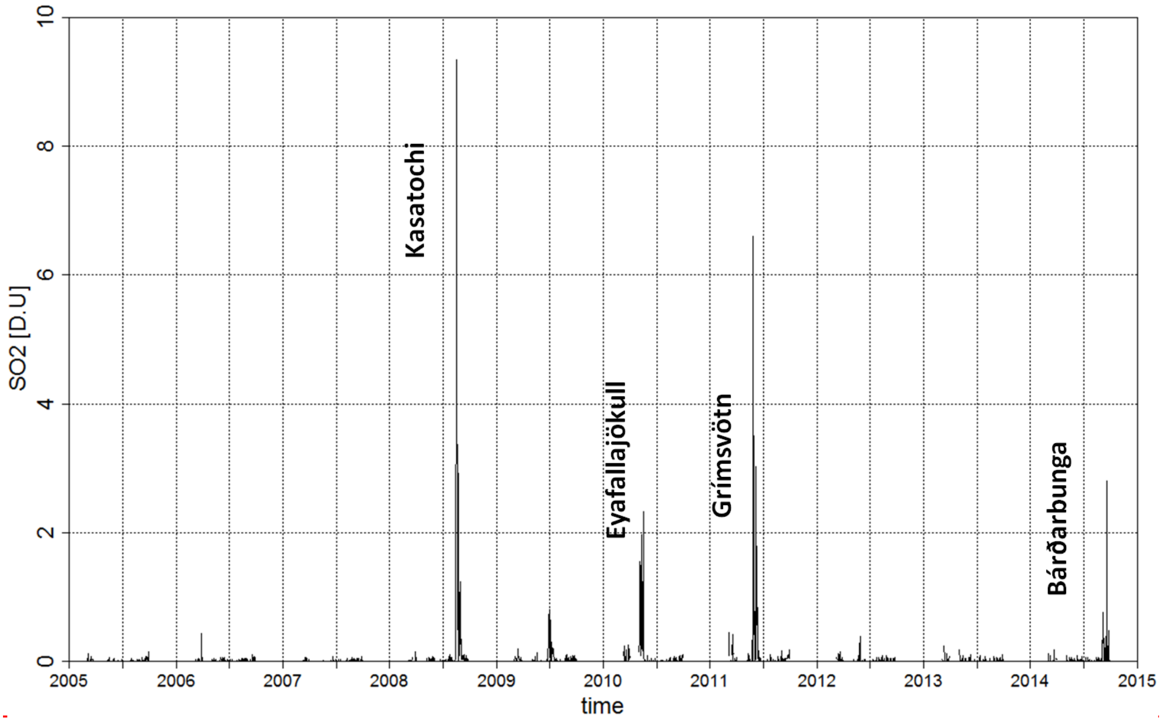
(*) missing values are those possessing < 25 days of data in each bimonthly period, or no data.

In brackets: Values exceeding ±3σ of the mean of all stations in each bimonthly period were not included in the analysis.

~~**Table 5. Summary of correlation coefficients between the mean columnar SO_2 measured by the brewers in the Northern Hemisphere and provided by the satellite products of OMI and GOME-2 during the Kasatochi eruption in August 2008.**~~

	~~01/08/2008 – 31/08/2008~~
~~Brewers and GOME-2~~	~~0.936 (*)~~
~~Brewers and OMI (TRM)~~	~~0.893 (*)~~
~~Brewers and OMI (PBL)~~	~~0.809 (*)~~

~~(*) p-value < 0.0001~~



~~**Figure 1.** SO_2 column in the past decade as monitored over Iceland (60°N–70°N, 25°W–15°W) from OMI. Shown are peaks which can be attributed to four volcanoes detected by OMI and collocated Brewer instruments.~~

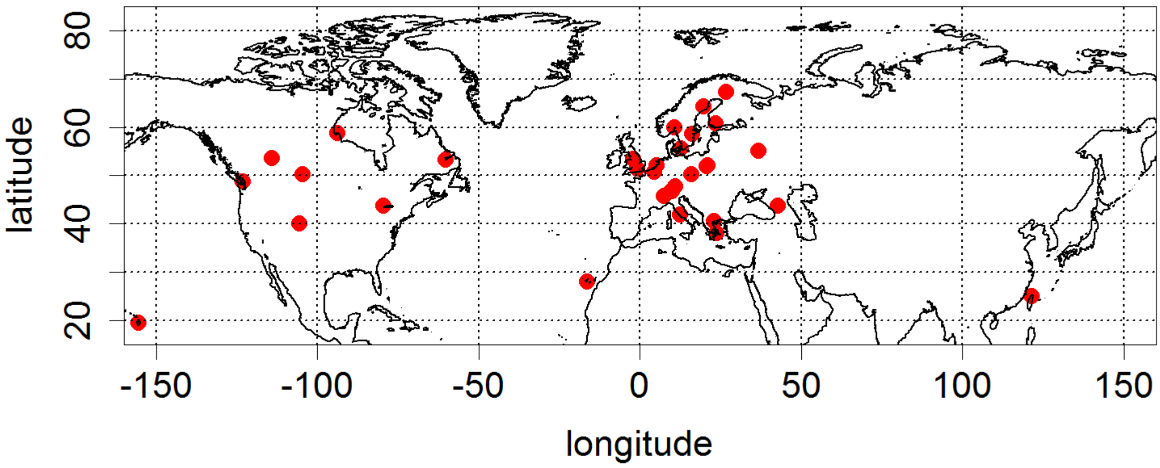
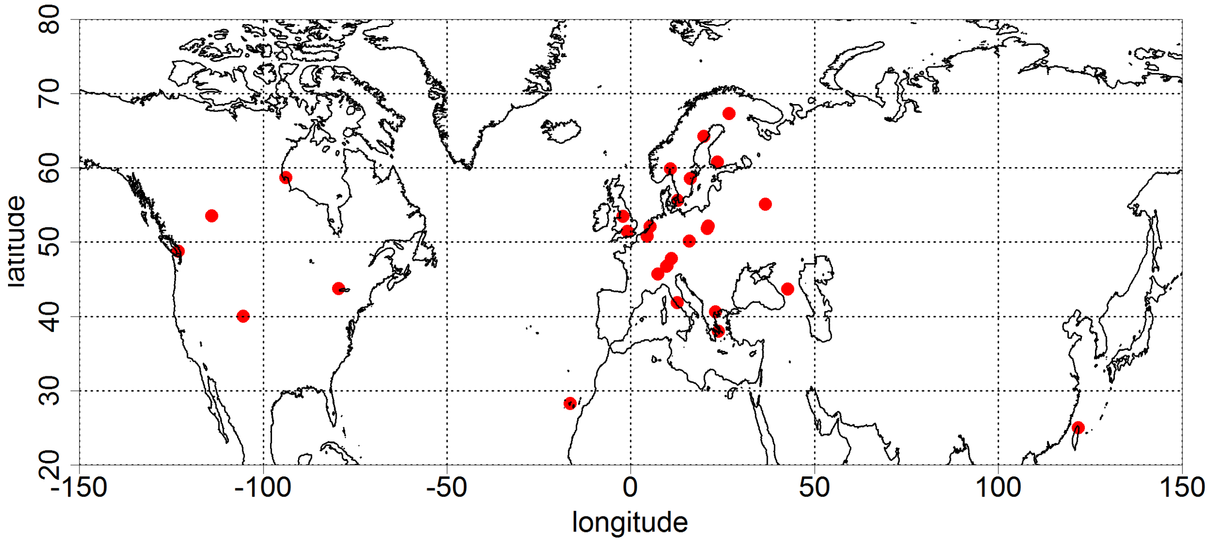


Figure 1. All stations with accessible SO_2 column data from Brewers analysed in this study as listed in Table 2.



~~**Figure 2. All stations with accessible SO_2 column data from Brewers analysed in this study.**~~

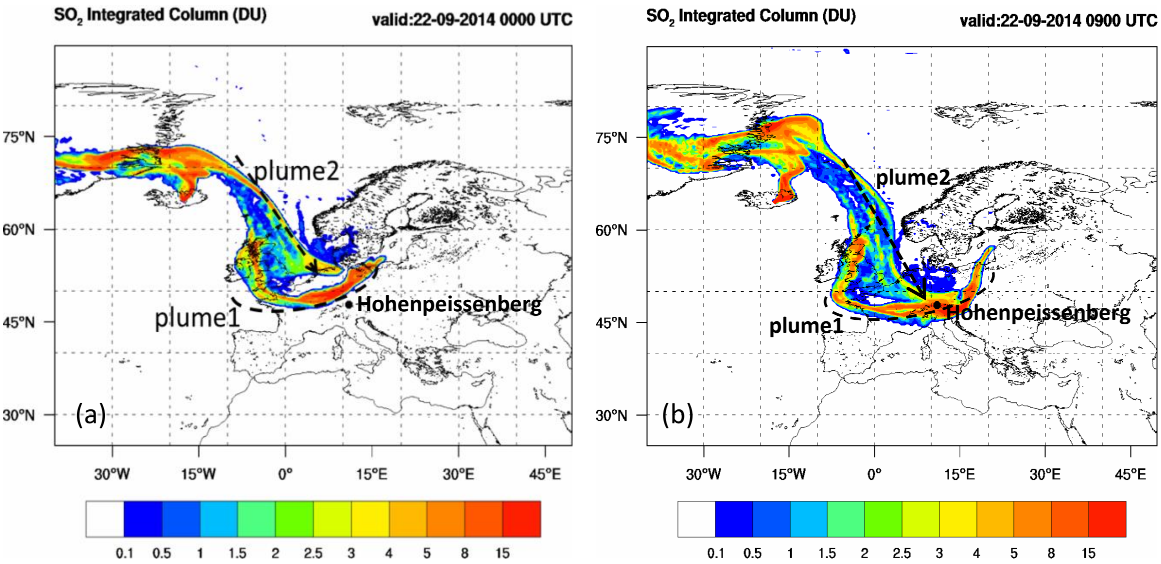


Figure ~~3~~2. Integrated column of SO_2 (DU) from Bárðarbunga emissions as simulated with FLEXPART-WRF model, a) 22 September 2014 00:00 UTC; b) 22 September 09:00 UTC. Dashed lines indicate the orientation of the two distinct plumes overlapping over central Europe.

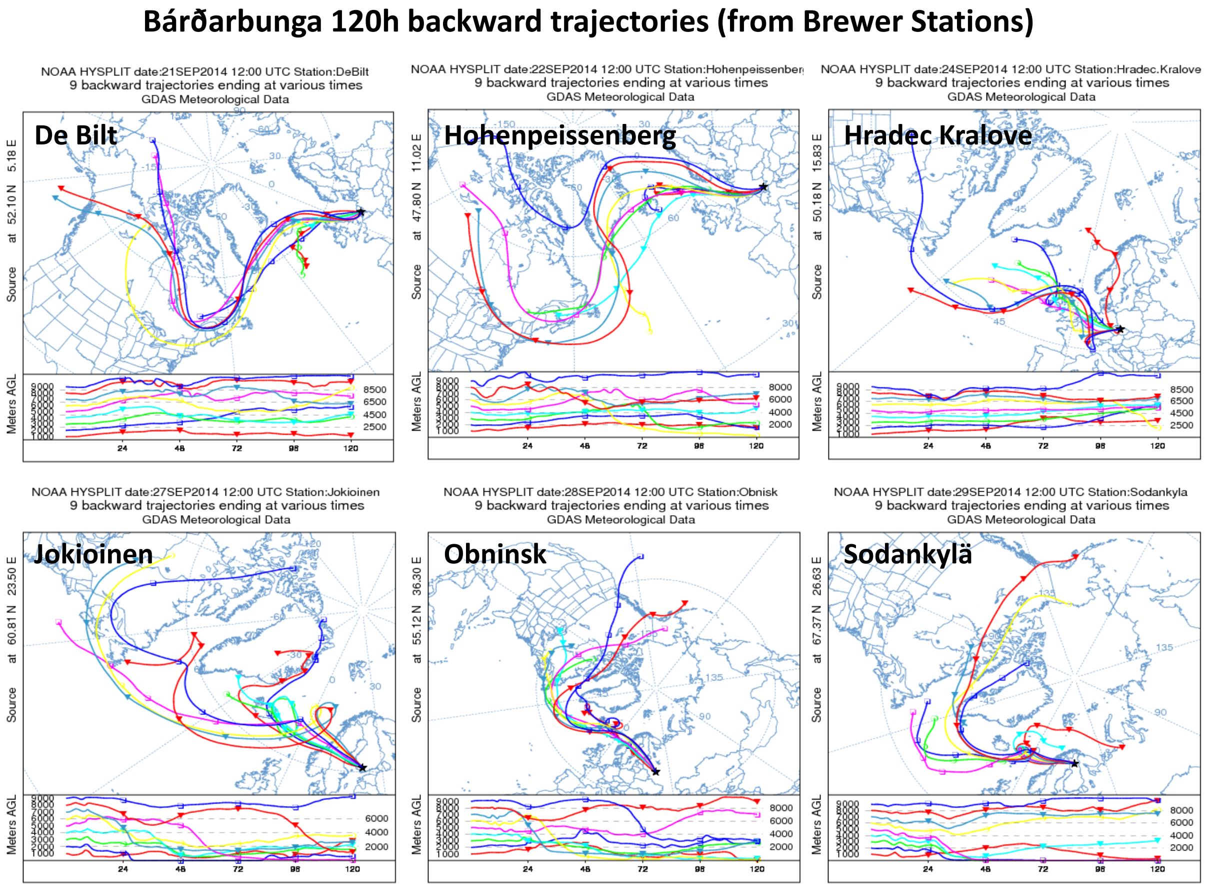


Figure 43. HYSPLIT 120 hours back trajectories of air masses arriving on the day of maximum SO_2 records for each one of the Brewer stations at De Bilt, Hohenpeissenberg, Hradec Kralove, Jokioinen, Obninsk and Sodankylä.

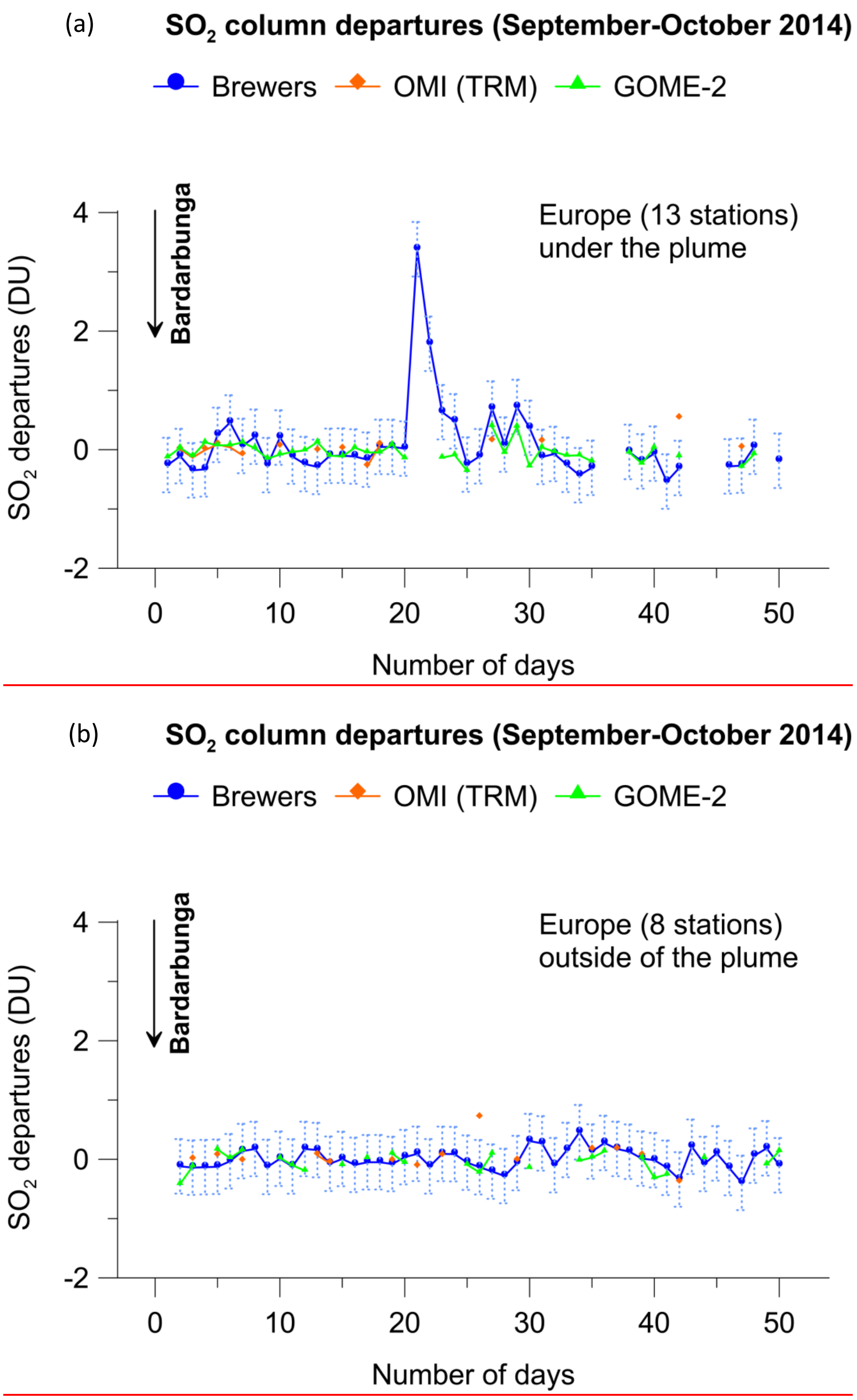
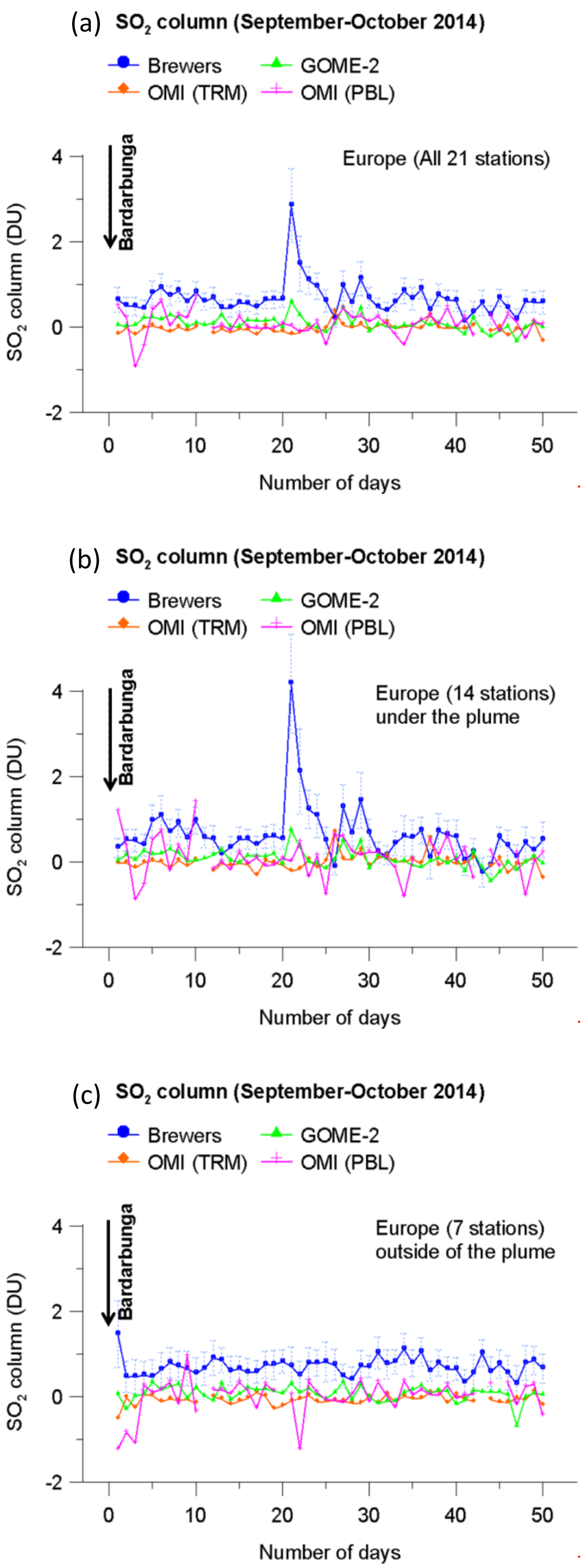


Figure 4. Mean SO_2 column departures from the unperturbed 10 day pre-volcanic baseline measured by Brewers, OMI (TRM) and GOME-2 during September-October 2014 over Europe following the 2014 Bárðarbunga volcanic eruption for: (a) stations under the volcanic SO_2 plume, and (b) stations outside of the plume. The error bars for the Brewer observations show the standard deviation of all daily values during the unperturbed 10 day period prior to the volcanic eruption. Brewer stations under the plume are: Sodankÿla, Vindeln, Jokioinen, Oslo, Norrkoeping, Copenhagen, Obninsk, Manchester, De Bilt, Reading, Uccle, Hradec Kralove, Hohenpeissenberg and Aosta. Stations outside of the plume are: Warsaw, Belsk, Davos, Arosa, Kislovodsk, Rome and Athens. Each daily average from either OMI or GOME-2 was calculated if and only if more than half of the individual overpasses had data at a given day.



~~**Figure 5. (a) Mean SO_2 column in DU measured by Brewers, OMI and GOME-2 during September-October 2014 over Europe. (b) Same as (a) but for stations under the plume. (c) Same as (a) but for stations outside the plume. The error bars for the Brewer observations show the standard error of all daily values entering the average.**~~

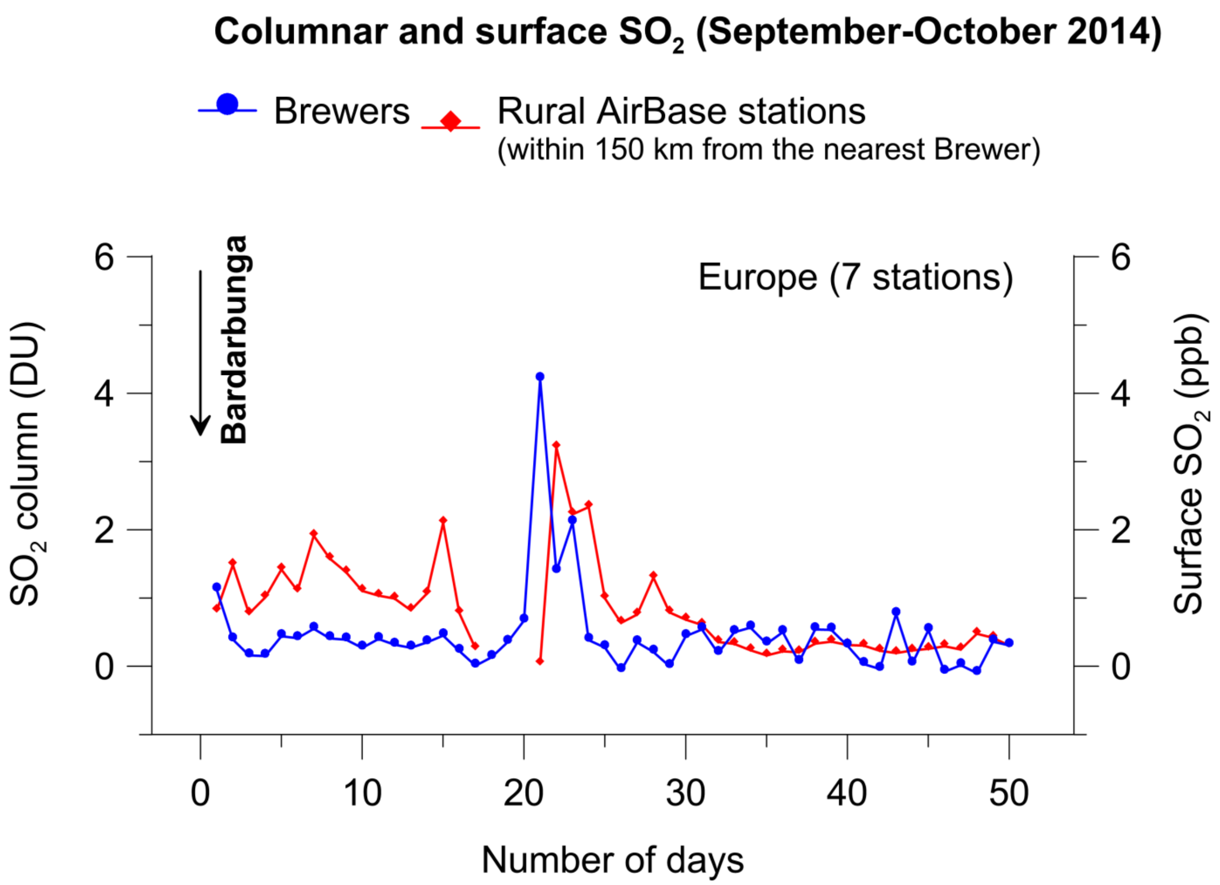


Figure 5. Mean surface SO_2 measured by Airbase class 1-2 stations located within 150 km from 7 nearest Brewer stations in Europe as listed in Table 3.

~~**Figure 6. Mean surface SO_2 measured by Airbase class 1-2 stations located within 150 km from the nearest Brewer station.**~~

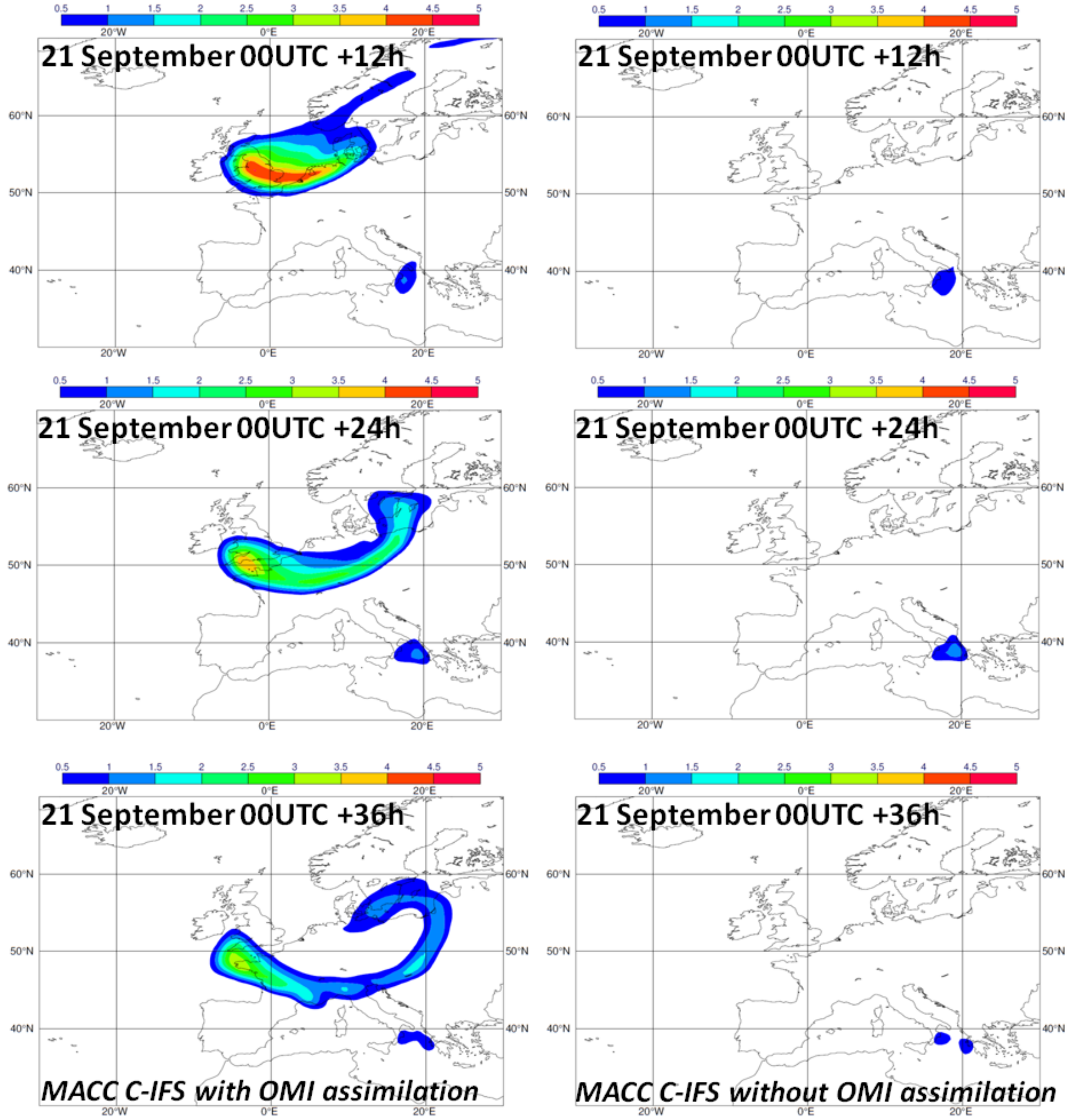


Figure ~~7~~6. Charts of forecasted total column SO_2 produced within the MACC system for 21 September 2014 with OMI data assimilation (left) and without OMI data assimilation (right).

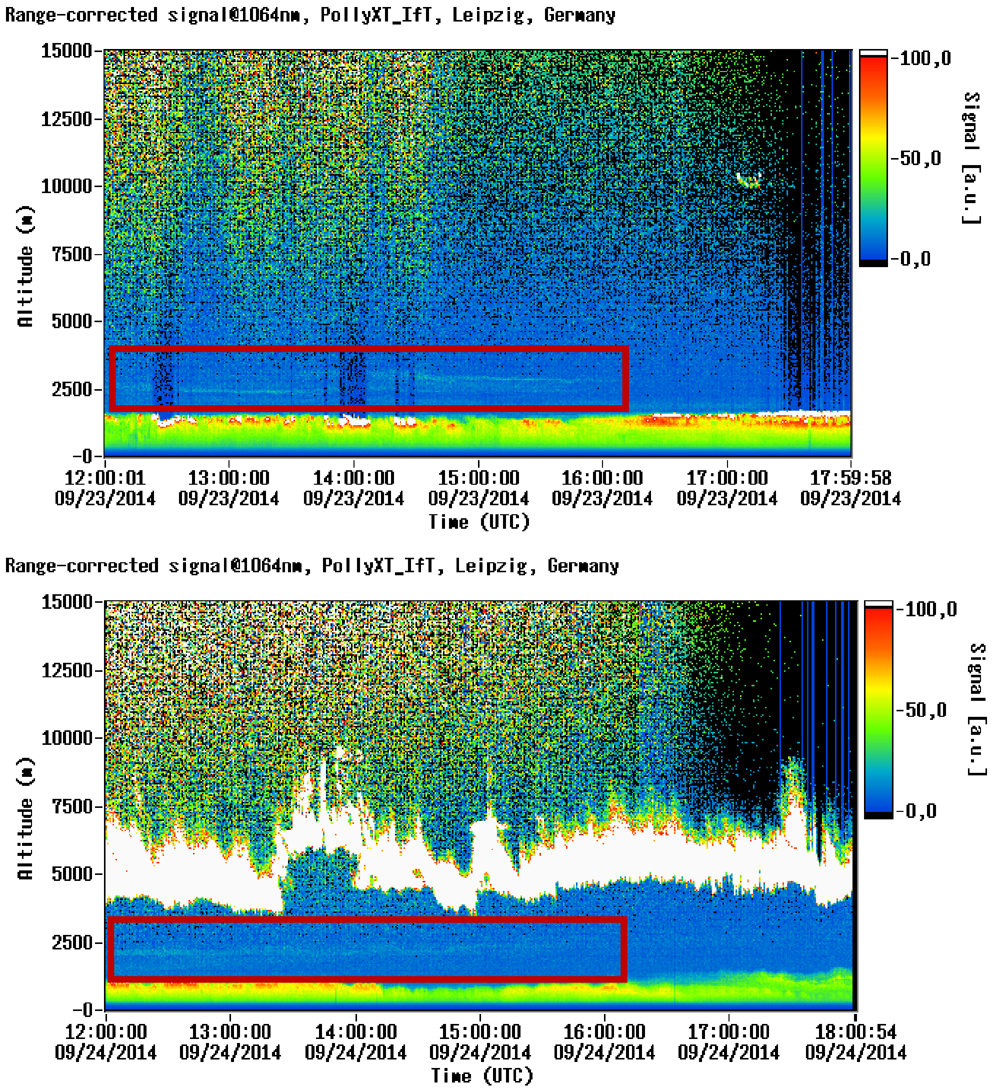
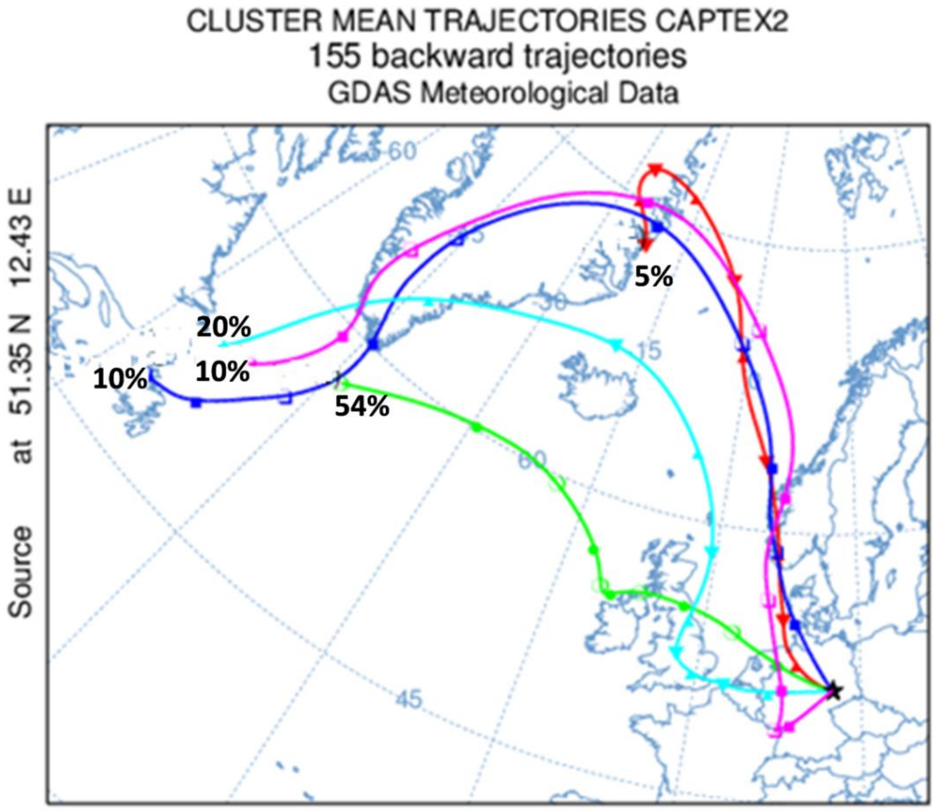


Figure ~~8~~7. Range corrected signal at 1064 nm from the PollyXT lidar in Leipzig on 23 September 2014 (up) and 24 September 2014 (down). The red rectangular indicates the location of the volcanic ash layer.



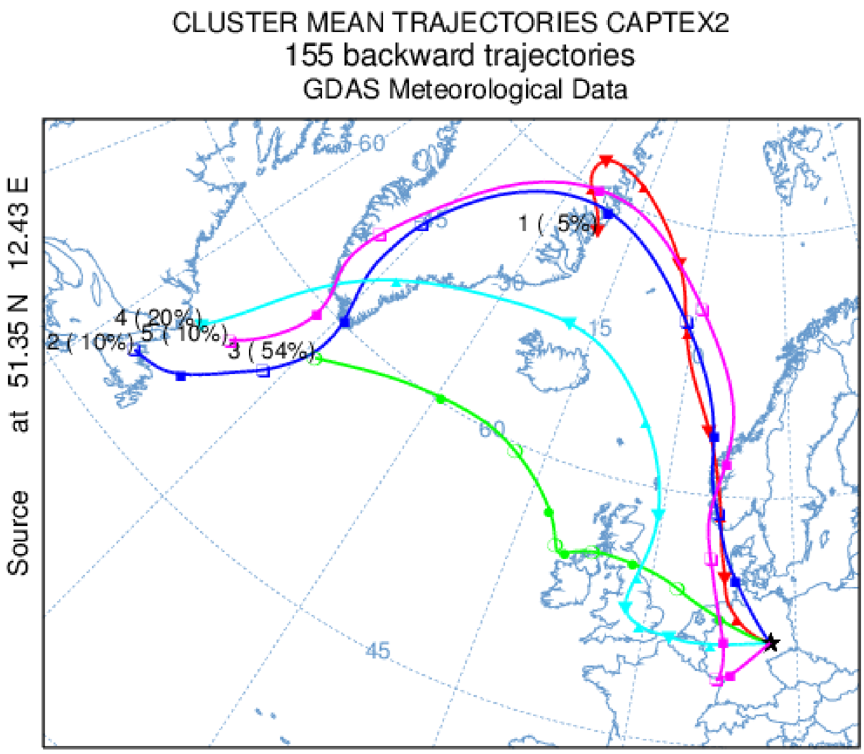


Figure ~~9~~8. Cluster analysis of the HYSPLIT back trajectories that arrive every hour (from 23 September 12:00 UTC up to 24 September 18:00 UTC) at 2.5-3.5 km height over Leipzig. A 54% cluster percentage means that there is 54% chance that the SO_2 arriving anywhere between 2.5-3.5 km over Leipzig originates from the specific direction.

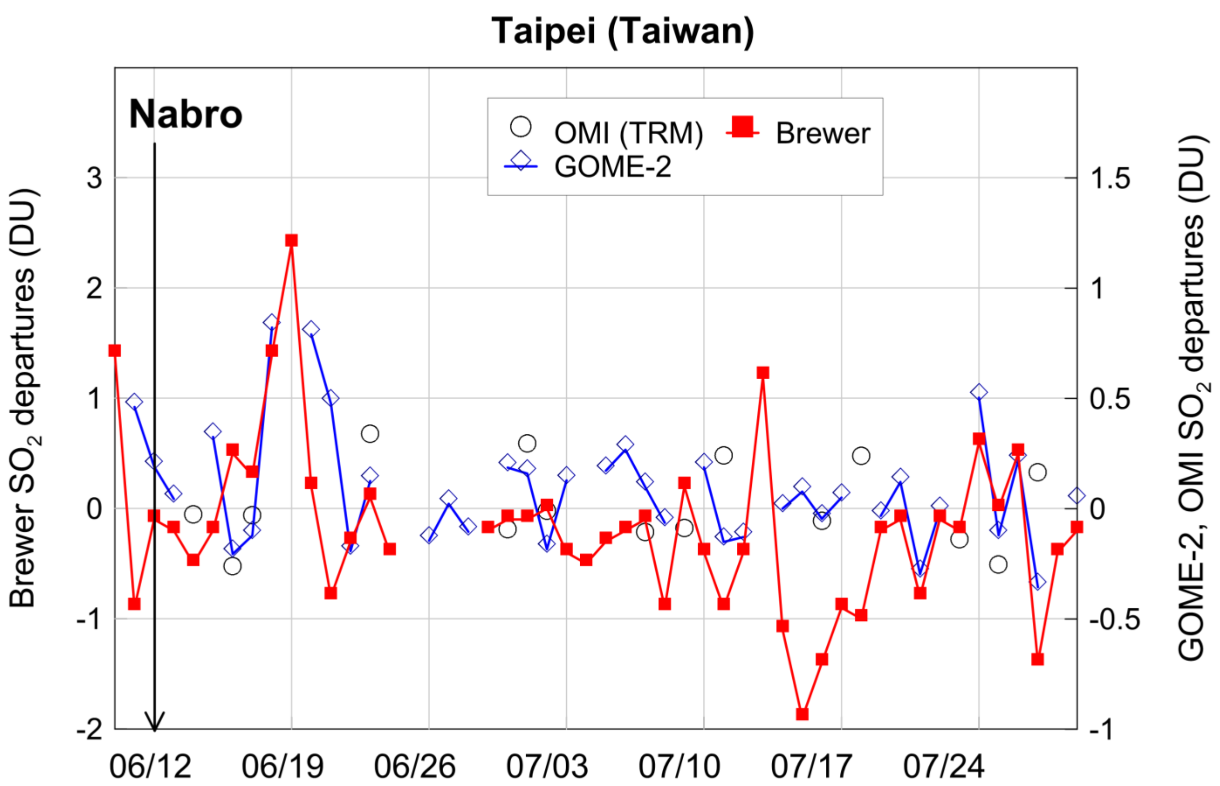
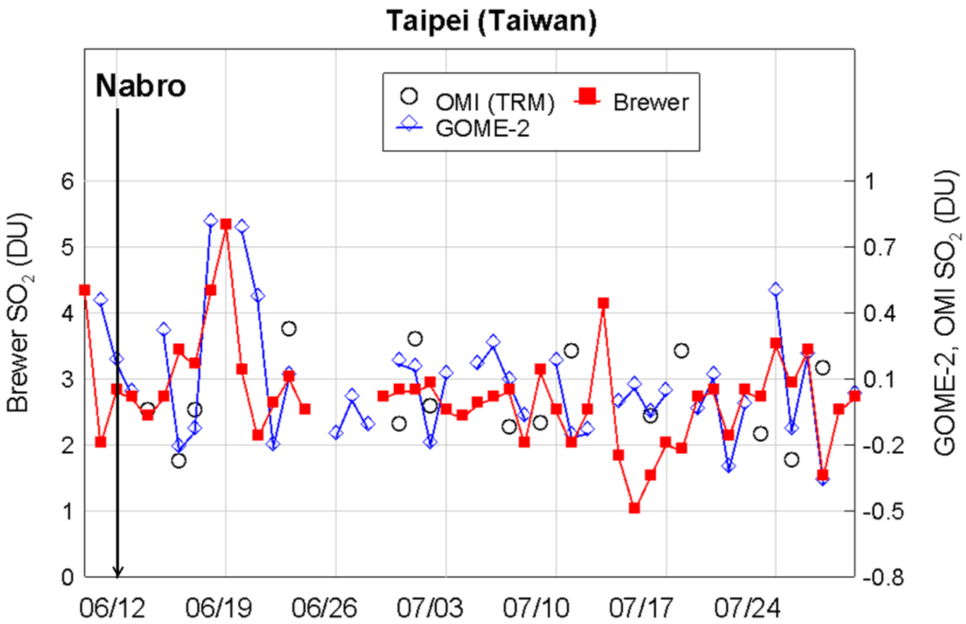


Figure 9. SO_2 column departures from the unperturbed 10 day pre-volcanic baseline measured by Brewer, OMI (TRM) and GOME-2 over Taipei, Taiwan, during June-July 2011 following the 2011 Nabro volcanic eruption.



~~**Figure 10. SO_2 columns from Brewer, OMI (TRM) and GOME-2 overpasses over Taipei, Taiwan, during June-July 2011.**~~

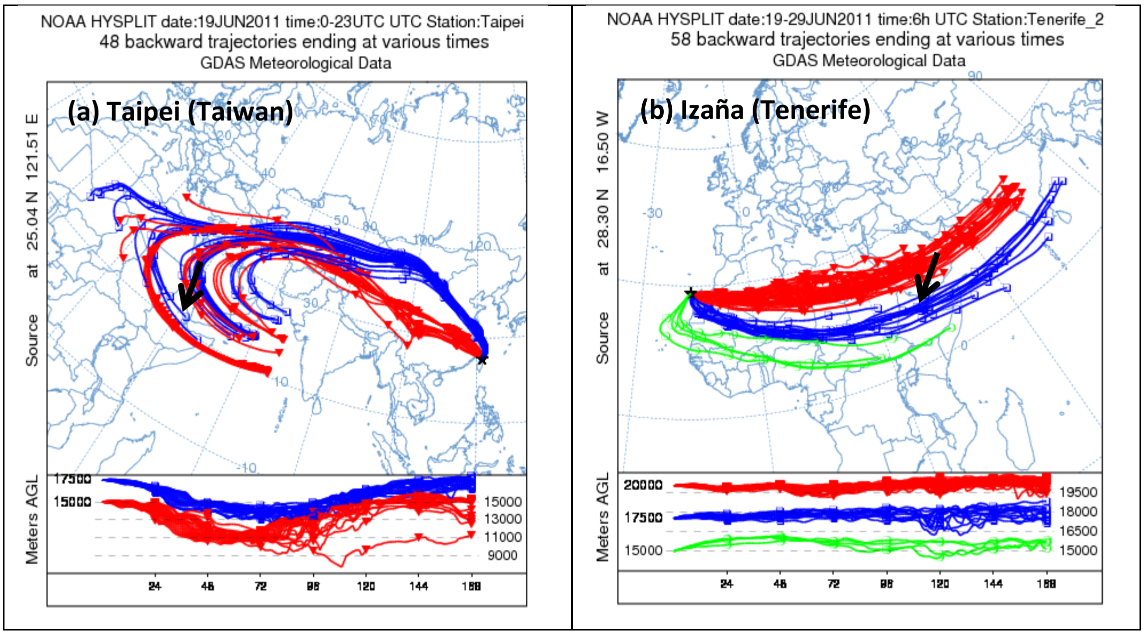


Figure ~~11~~10. HYSPLIT back trajectories of air masses (a) from Taipei (Taiwan) on 19 June 2011, (b) from Izaña (Tenerife) for days 19-29 June 2011. Nabro's location is indicated by the black arrow.

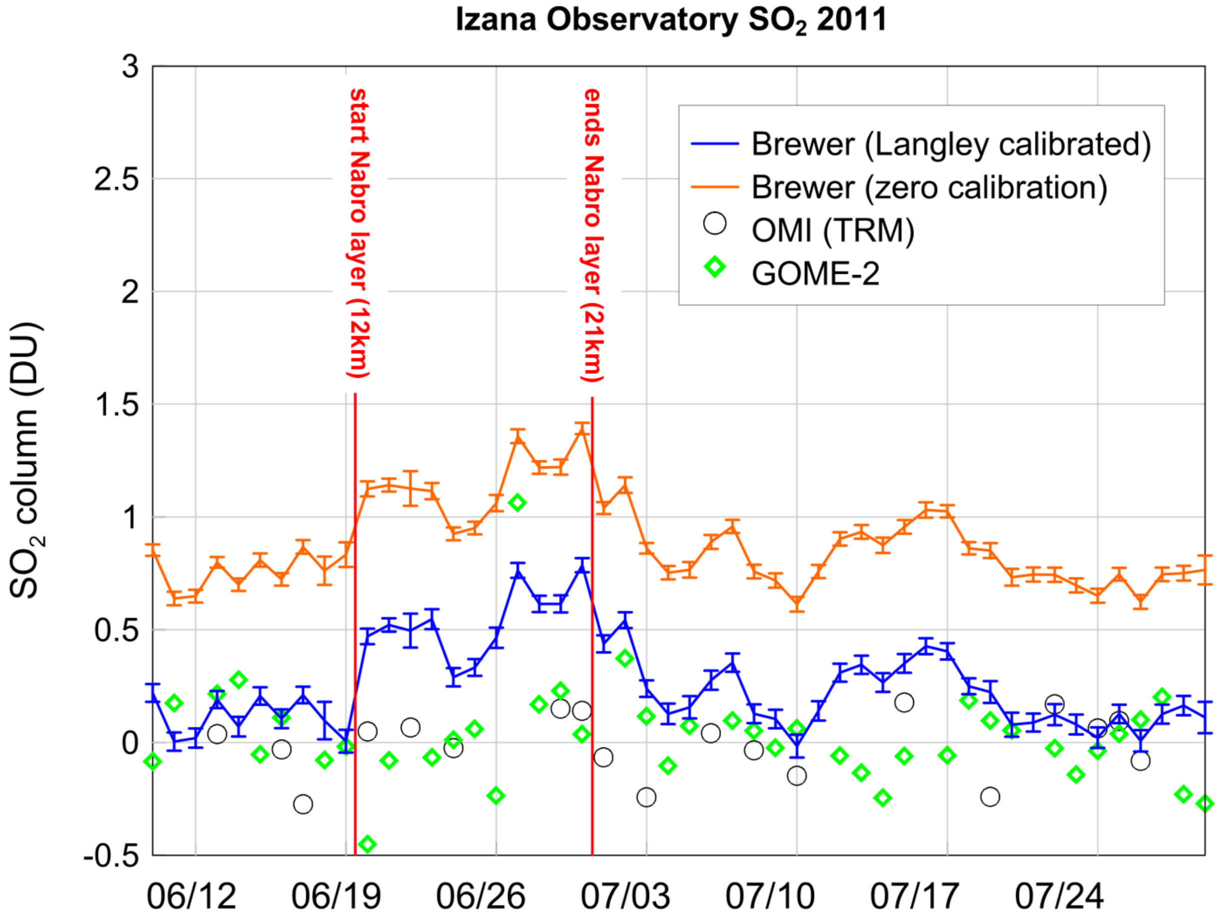
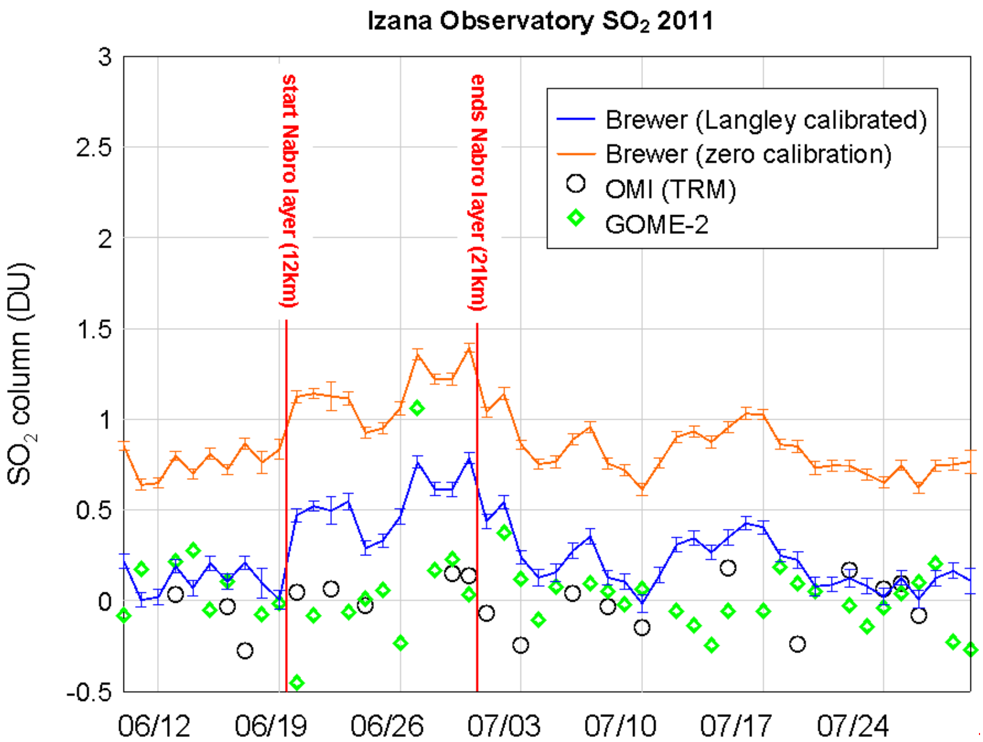
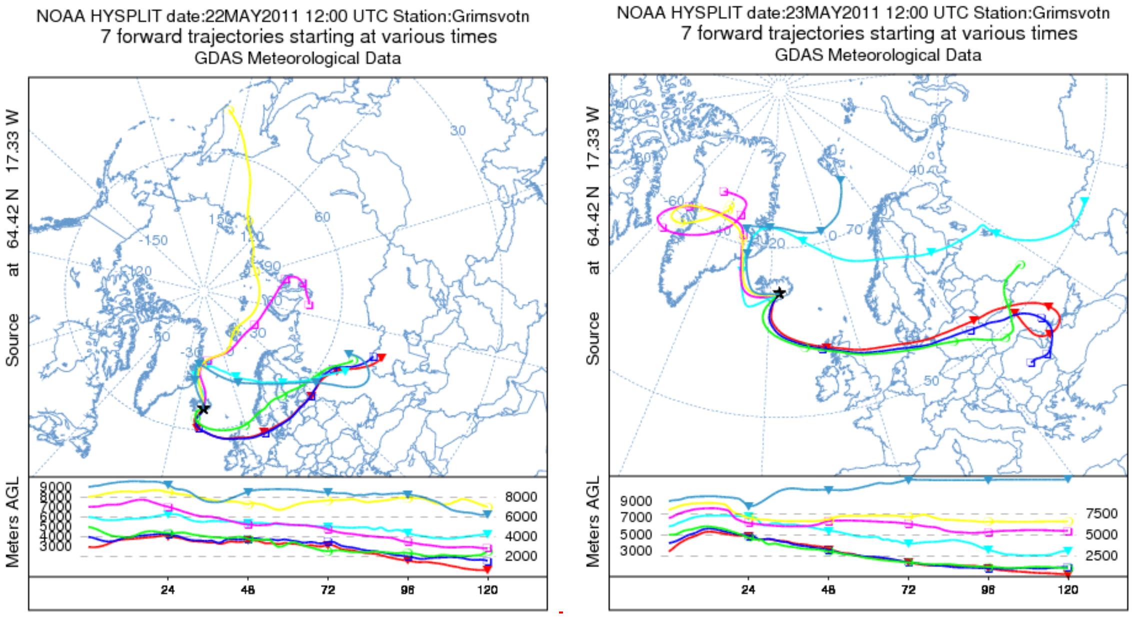


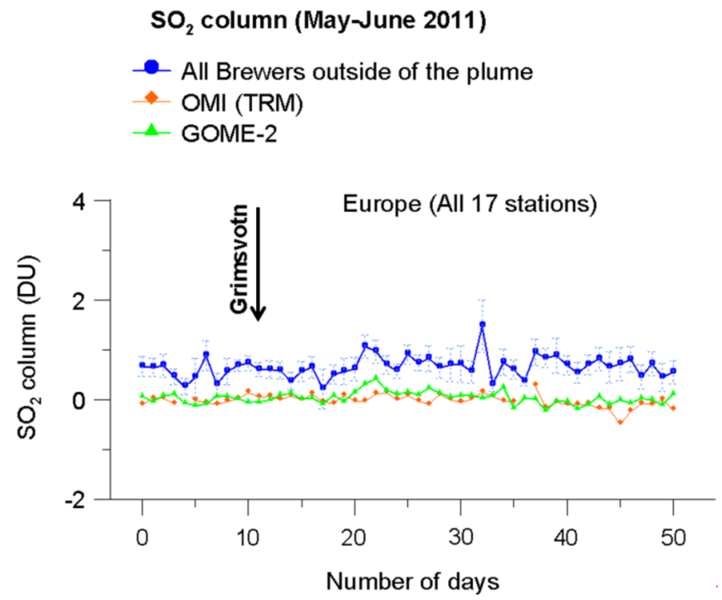
Figure 11. SO_2 calculations using the Langley calibration and the zero calibration at Izaña (assuming SO_2=0 during the days 06 and 07 of June 2011) following the 2011 Nabro volcanic eruption. Also shown are SO_2 columns from OMI (TRM) and GOME-2 overpasses over Izaña Observatory during June-July 2011.



~~**Figure 12. SO_2 calculations using the Langley calibration and the zero calibration (assuming SO_2=0 during the days 06 and 07 of June 2011). Also shown are SO_2 columns from OMI (TRM) and GOME-2 overpasses over Izaña Observatory during June-July 2011.**~~



~~Figure 13. HYSPLIT 120 hours forward trajectories from Iceland following Grímsvötn eruption.~~



~~**Figure 14. All Brewer stations were outside of the Grímsvötn volcanic eruption plume. The error bars for the Brewer observations show the standard error of all daily values entering the average.**~~

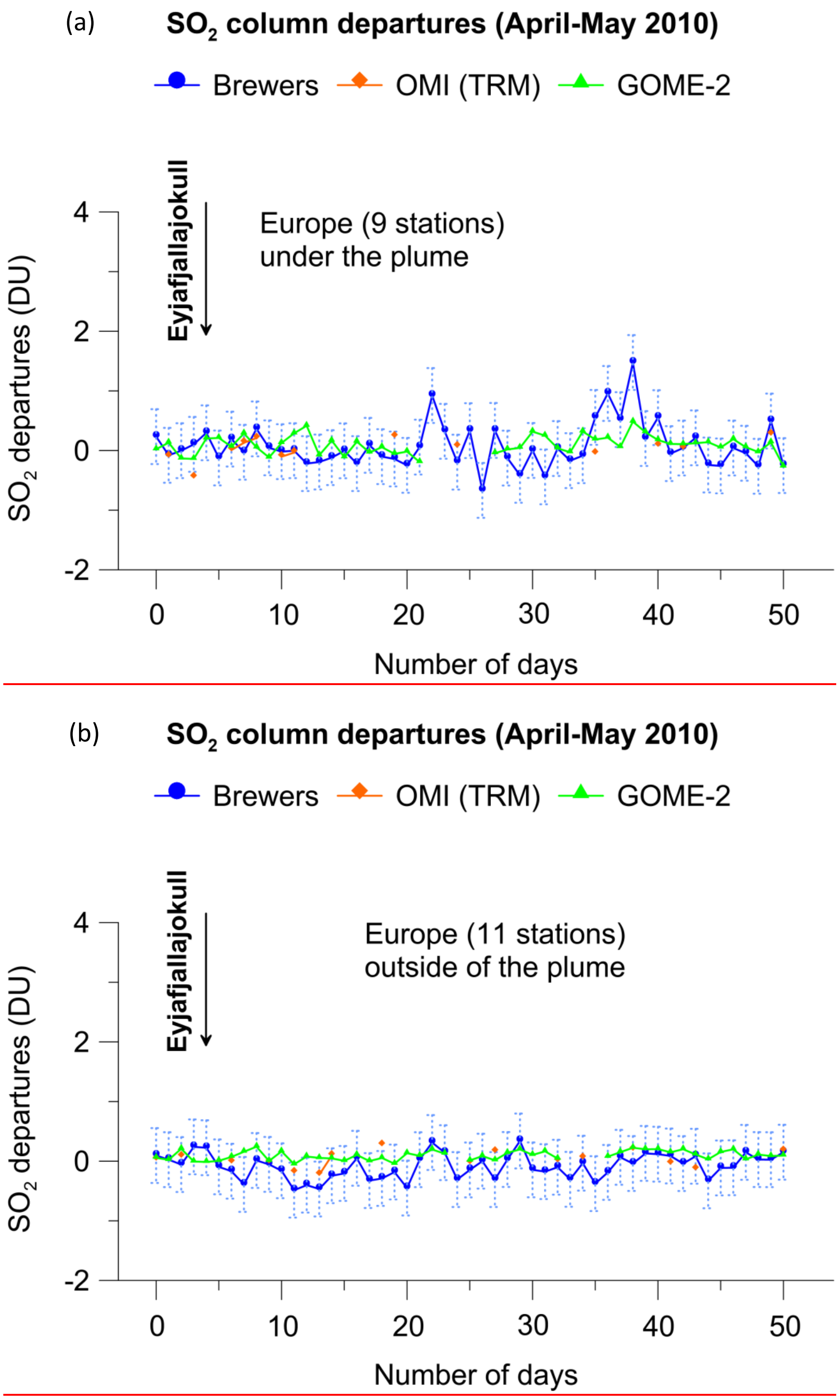
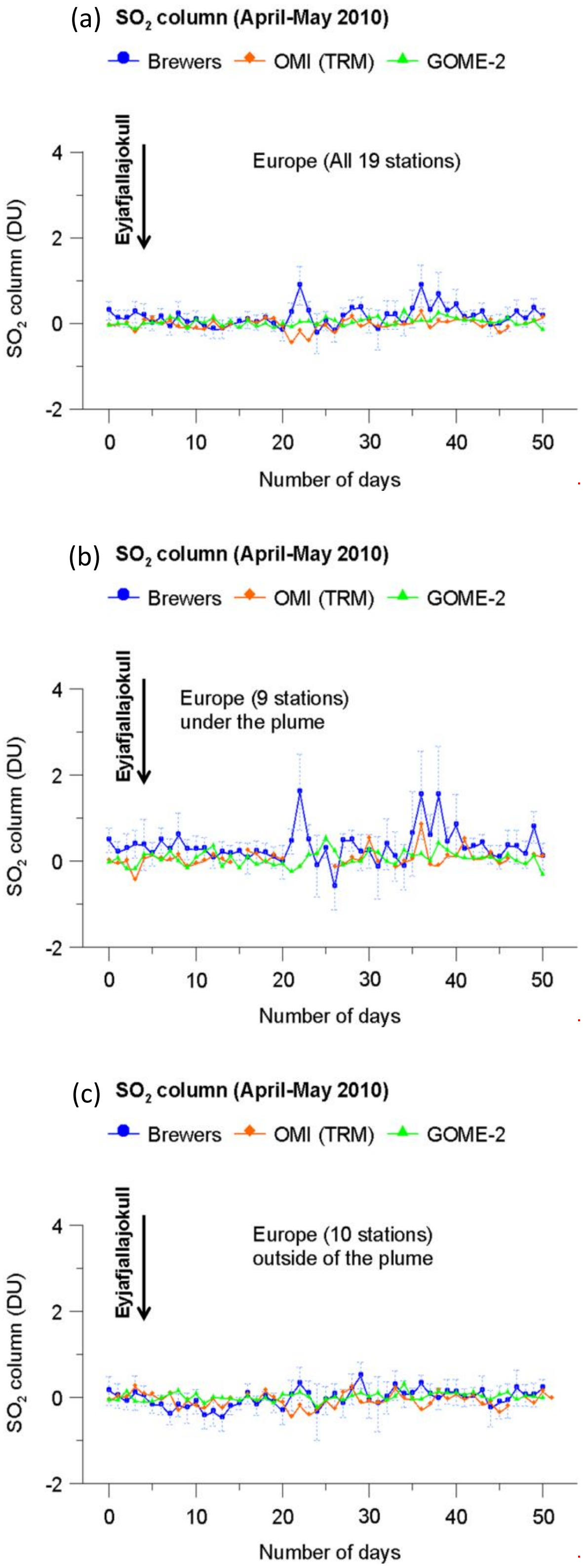


Figure 12. Mean SO_2 column departures from the unperturbed 10 day pre-volcanic baseline measured by Brewers, OMI (TRM) and GOME-2 during April-May 2010 over Europe following the 2010 Eyjafjallajökull volcanic eruption for: (a) stations under the volcanic SO_2 plume, and (b) stations outside of the plume. The error bars for the Brewer observations show the standard deviation of all daily values during the unperturbed 10 day period prior to the volcanic eruption. Brewer stations under the plume are: Sodankÿla, Obninsk, Manchester, De Bilt, Belsk, Reading, Hohenpeissenberg, Davos and Arosa. Stations outside of the plume are: Vindeln, Oslo, Norrkoeping, Copenhagen, Uccle, Hradec Kralove, Aosta, Kislovodsk, Rome, Thessaloniki and Athens. Each daily average from either OMI or GOME-2 was calculated if and only if more than half of the individual overpasses had data at a given day.



~~**Figure 15. (a) Mean SO_2 column measured by Brewers, OMI and GOME-2 during April-May 2010. (b) Same as (a) but for stations under the plume. (c) Same as (a) but for stations outside of the plume. The error bars for the Brewer observations show the standard error of all daily values entering the average.**~~

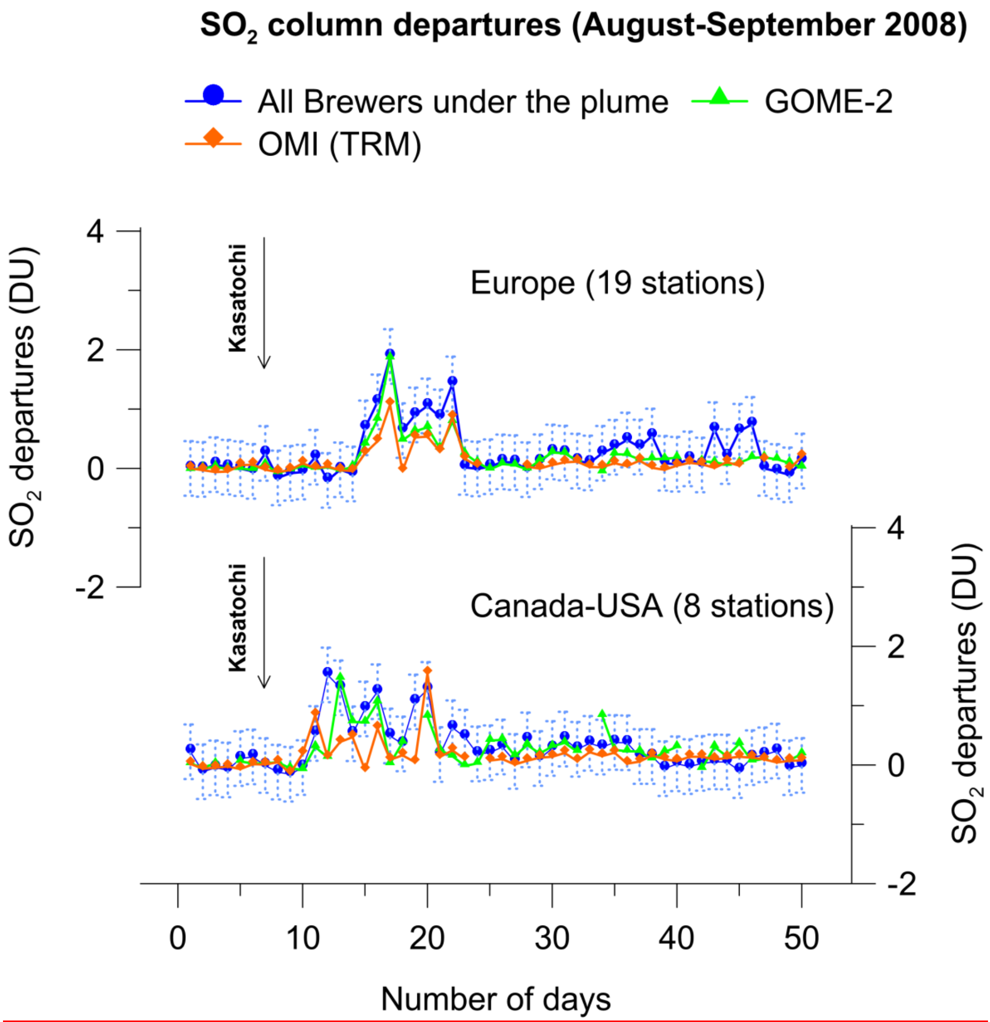
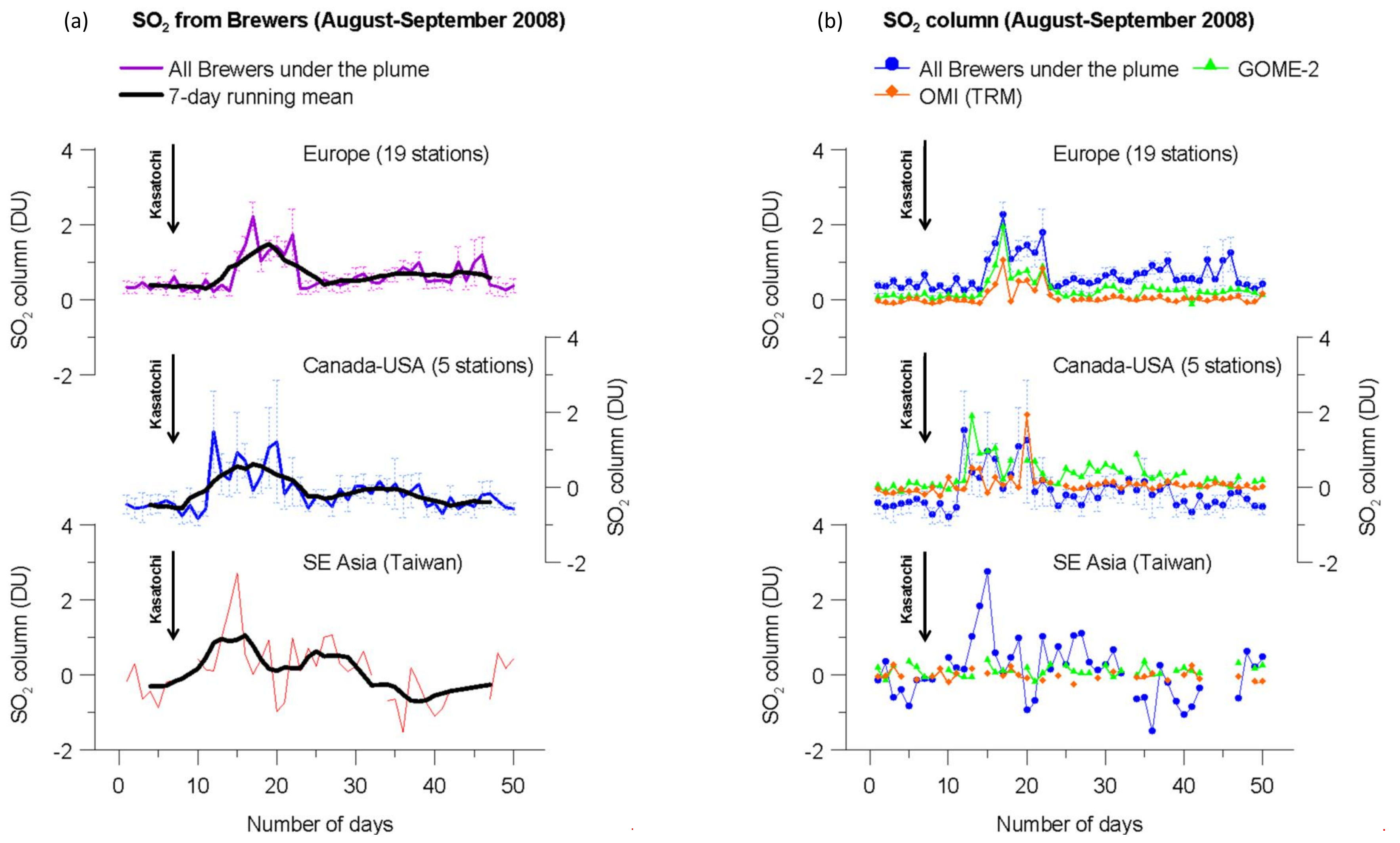


Figure 13. Mean SO_2 column departures from the unperturbed 10 day pre-volcanic baseline measured by Brewers, OMI (TRM) and GOME-2 during August-September 2008 over Europe and Canada/USA following the 2008 Kasatochi volcanic eruption. The error bars for the Brewer observations show the standard deviation of all daily values during the unperturbed 10 day period prior to the volcanic eruption. Stations in Europe include: Sodankÿla, Vindeln, Jokioinen, Norrkoeping, Copenhagen, Manchester, De Bilt, Belsk, Reading, Uccle, Hradec Kralove, Hohenpeissenberg, Davos, Arosa, Aosta, Kislovodsk, Rome, Thessaloniki and Athens. Stations in Canada/USA include: Churchill, Edmonton, Goose Bay, Regina, Saturna Island, Toronto, Boulder and Mauna Loa. Each daily average from either OMI or GOME-2 was calculated if and only if more than half of the individual overpasses had data at a given day.



~~**Figure 16. Mean SO_2 column measured by Brewers, OMI (TRM) and GOME-2 over Europe, Canada/USA and Taiwan during August-September 2008. The error bars for the Brewer observations show the standard error of all daily values entering the average.**~~