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.

<u>Answers to specific comments</u>

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:

<u>Answers to main comments</u>

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.