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

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Abstract. 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

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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 project shows that the large increases of SO_2 over Europe following the Bárðarbunga eruption in Iceland were not caused by local sources or ship emissions but are clearly linked to the eruption. 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 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) increases of column SO_2 amounts unless observations are made near a source. 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 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 focusing mostly on aviation, e.g. ESA's Support to Aviation Control Service (SACS) (Brenot

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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. 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°N, 168.13°W, 1073 m above sea level (asl), 12 July 2008, Prata et al., 2010) and Sarychev, Russia (48.1°N, 153.2°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

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data. 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 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%.

The paper is structured in the following Sections: 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 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

SO_2 in the atmosphere can be measured from ground-based instruments, by instrumentation onboard the spacecraft and can be 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 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 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 levels (Koukouli et al., 2014). Details on the method with which SO_2 is measured with the Brewer spectrophotometer can be found in Kerr et al. (1980; 1985; 1988) and De Backer and De Muer (1991). 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 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.

In this study we analysed twenty three stations located in the European Union, four Brewer stations in Canada, one in the USA and one in Taiwan, whose geographical positions are shown in Figure 2. SO_2 measurements were averaged over a large number of instruments and datasets during periods following volcanic eruptions.

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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, Edmonton, Saturna Island, Toronto in Canada, and Taipei in Taiwan 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 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. We mention here that most of the European Brewer data providers participate in a recent EU COST Action (EUBREWNET, http://www.eubrewnet.org/cost1207/) aiming 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 ±6σ 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 ± 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 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 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 low latitude sites in the Northern Hemisphere, namely Izaña 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 level.

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2.2 Satellite Data

The columnar SO_2 records from remote sensing spectrophotometers over Europe, Canada, USA and Taiwan were compared with spaceborne measurements from a) the Ozone Monitoring Instrument (OMI) on-board EOS-Aura (e.g. Ialongo et al., 2015) and b) the Global Ozone Monitoring Experiment-2 (GOME-2) on board 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 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 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 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, 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. 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 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 ($\sigma/\sqrt{N}$) 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

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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 and boundary conditions for the WRF model are from the National Center for Environmental Prediction (NCEP) final analysis (FNL) dataset at 1°×1° resolution and the sea surface temperature (SST) is from the NCEP 1°×1° analysis. 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 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). On 22 September the primary plume (plume_1) becomes stagnant over the topographic barrier of the Alps (Figure 3b). The secondary plume is advected southwards by the intense northerly winds over the North Sea and the two plumes overlap at about 09:00-11:00 UTC. 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, 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 surface SO_2 observed over the Netherlands during a period of several days.

The high SO_2 concentrations that were recorded almost simultaneously at stations over Europe in various sites during the period 21-29 September 2014, are therefore 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. 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 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

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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.

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 ground level, in the coast of France, in the United Kingdom, the Netherlands and Germany between 21 and 25 September 2014. This all pointed towards an 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 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

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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). 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. 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 different altitudes and may follow different trajectories. 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 and 9 demonstrate that the same approach can be applied which could contribute towards an early warning synergistic tool as evidenced in the example of the Bárðarbunga case. The role of the Brewer stations in this system will be the early detection of SO_2 plumes as long as they arrive over continental areas and the triggering of the associated forecasting systems (models and networks).

3.2 The 2011 Nabro Volcano plume

A major eruption of Mt. Nabro, a 2218m high volcano on the border between Eritrea and Ethiopia (13.37 ^{o}N, 41.7 ^{o}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 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). SO_2 signals of volcanic origin were detected both by Brewer and satellite measurements over eastern Asia where the volcanic SO_2 plume was transported, as can be seen from Figure 10 and Figure 11a. Measurements come from 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. 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.

Although the Nabro volcanic plume was mainly transported to the east into Asia and was detected by various

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satellite instruments which provide better spatial coverage than the Brewers, we present here an interesting case where an increase in the SO_2 column due to the volcanic SO_2 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 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 June 19th to 21 km on June 29th (Sawamura et al., 2012). The daily mean SO_2 record (Figure 12) shows a 0.5 DU increase at the beginning of the event (June 19th), reaching 0.75 DU on 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).

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 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 19/06 and 01/07 as it was the case with the Brewer instrument (Figure 12). During some days between 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, 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 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^{o}N, 17.33^{o}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

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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 The case of the 2010 Eyjafjallajökull volcanic eruption

The Eyjafjallajökull volcano, Iceland (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 shows the responses of Brewer stations under the 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 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 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 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 the examined Brewer stations were determined to be either under or outside of the volcanic SO_2 plume according to careful analysis of the 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.

As we can see from Figure 15, the 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 SO_2 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 that the average SO_2 columns measured by the Brewers under the plume during these three periods 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 measured by the Brewers under the plume during these two periods were estimated to 0.1 ± 0.4 DU and 1.0 ± 0.5 DU respectively, both within the error

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bars. These amounts were higher than the amounts measured outside of the plume (-0.2 ± 0.3 DU and 0.2 ± 0.2 DU, accordingly) almost by 0.5 DU on average.

3.5 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) 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 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) and it was also detected by the majority of the Brewers in Europe with a delay of about 3 days. The total SO_2 column averaged over Canada during the period 12-20 August 2008 is estimated to 0.8 ± 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 column of 1.4 ± 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 gives a rough estimate of the average volcanic SO_2 column measured by the Brewers over Europe. 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 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. The correlation between SO_2 from the Brewers and SO_2 from GOME-2 at 19 stations averaged over Europe is +0.933 (t-value = 13.98235, p-value < 0.0001, N = 31). Accordingly, the correlation between Brewer and OMI (TRM) SO_2 data is +0.919 (t-value = 12.34644, $p < 0.0001$, N = 30) and between GOME-2 and OMI (TRM) data is +0.922 (t-value = 12.63061, $p < 0.0001$, N = 30). These correlations were calculated from 30 daily averages during the Kasatochi volcanic eruption in August 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 three datasets 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 and provided by the satellite products of OMI and GOME-2 during the globally important Kasatochi event. The correlation coefficients have high statistical significance explaining more than 80% of the total variance between the columnar SO_2 measurements from ground and space.

4 Conclusions

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. 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 modeling tool could be proposed as follows: a modeling 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) or when a lidar instrument detects highly depolarizing layers that were not advected from a desert. The operational use of such a synergistic activity could provide near-to-real time and forecasting information on the evolution of volcanic episodes and also develop a comprehensive database of measurements useful to improve model results. This new well-tuned and organized 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.

5 Data availability

SO_2 columns at Churchill, Edmonton, Saturna Island, Toronto in Canada, and Taipei in Taiwan were obtained from the World Ozone and Ultraviolet Radiation Data Centre (WOUDC; http://www.woudc.org/). SO_2 columns at Niwot Ridge, USA, were downloaded from the NOAA-EPA Brewer Spectrophotometer UV and Ozone Network (NEUBrew; http://www.esrl.noaa.gov/gmd/grad/neubrew/). OMI and GOME-2 satellite SO_2 data products were downloaded from the Aura Validation Data Center (AVDC) at the website http://avdc.gsfc.nasa.gov/index.php?site=245276100. Surface SO_2 concentrations over Europe were acquired

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from the European Environment Agency databases (AirBase) http://www.eea.europa.eu/data-and-maps/data/aqereporting-1#tab-european-data.

Appendix A

Table A1. Dates where 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 considered to be under the 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
Norrkoeping	58.58	16.15	43	Outside the plume	30/9
Copenhagen	55.63	12.67	50	Outside the plume	26/9
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	29/9
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
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. 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. 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

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

		August-September 2008			April-May 2010			May-June 2011			September-October 2014		
(a)	Latitude	mean	σ	N (days)	mean	σ	N (days)	mean	σ	N (days)	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.

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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

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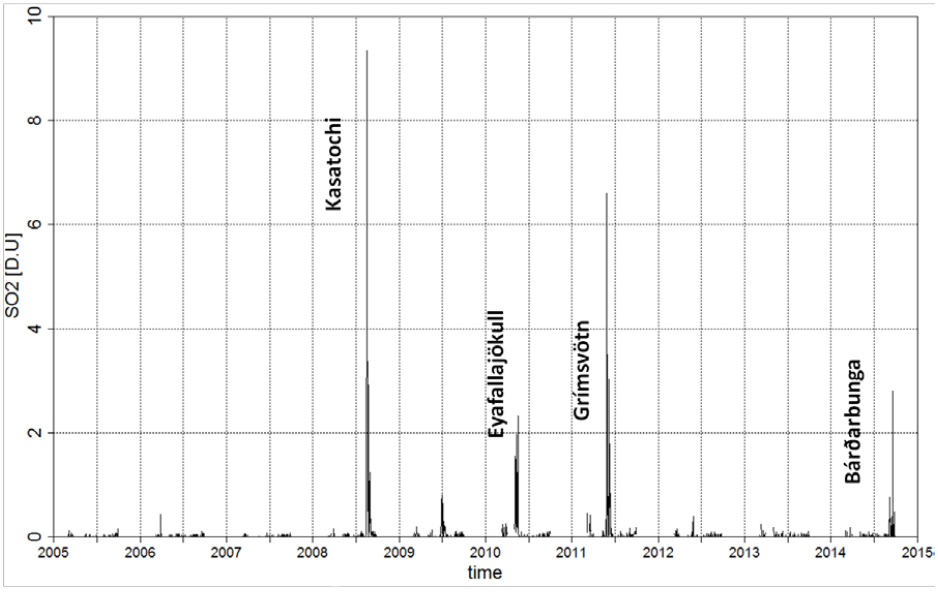

Figure 1. SO_2 column in the past decade as monitored over Iceland (60°N–70°N, 25°W–15°W) from OMI. Shown are peaks which can be attributed to four volcanoes detected by OMI and collocated Brewer instruments.

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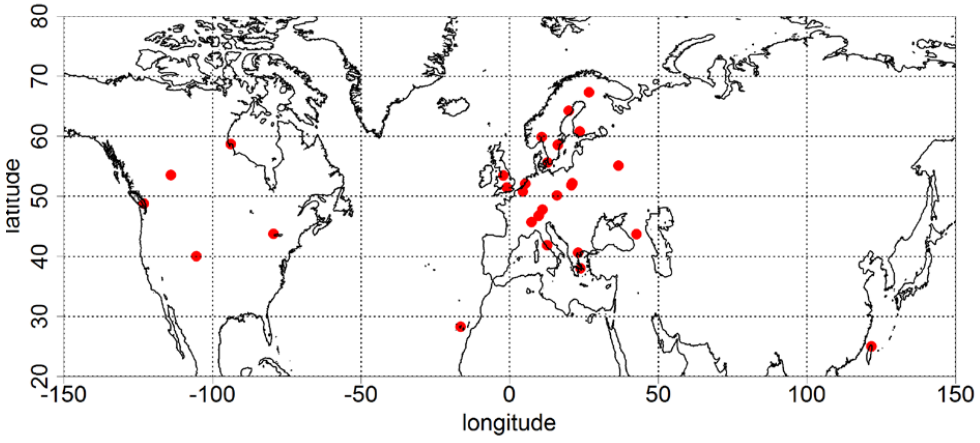

Figure 2. All stations with accessible SO_2 column data from Brewers analysed in this study.

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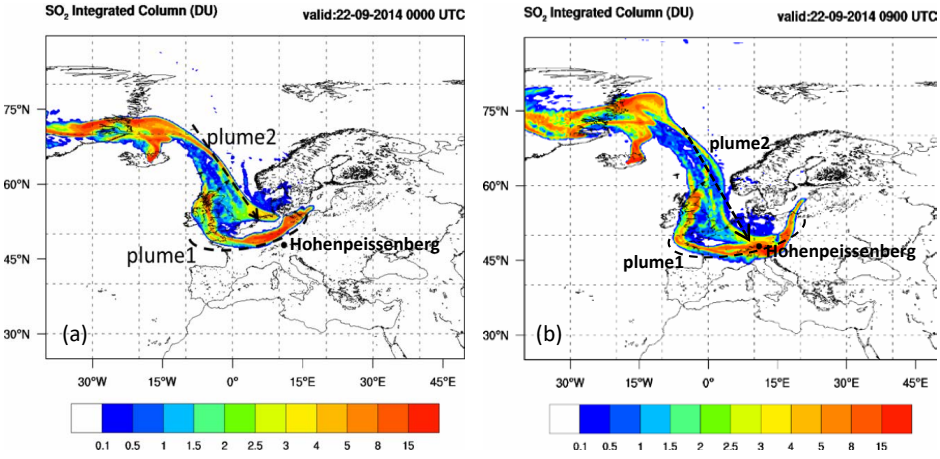

Figure 3. Integrated column of SO_2 (DU) from Bárðarbunga emissions as simulated with FLEXPART-WRF model, a) 22 September 2014 00:00 UTC; b) 22 September 09:00 UTC. Dashed lines indicate the orientation of the two distinct plumes overlapping over central Europe.

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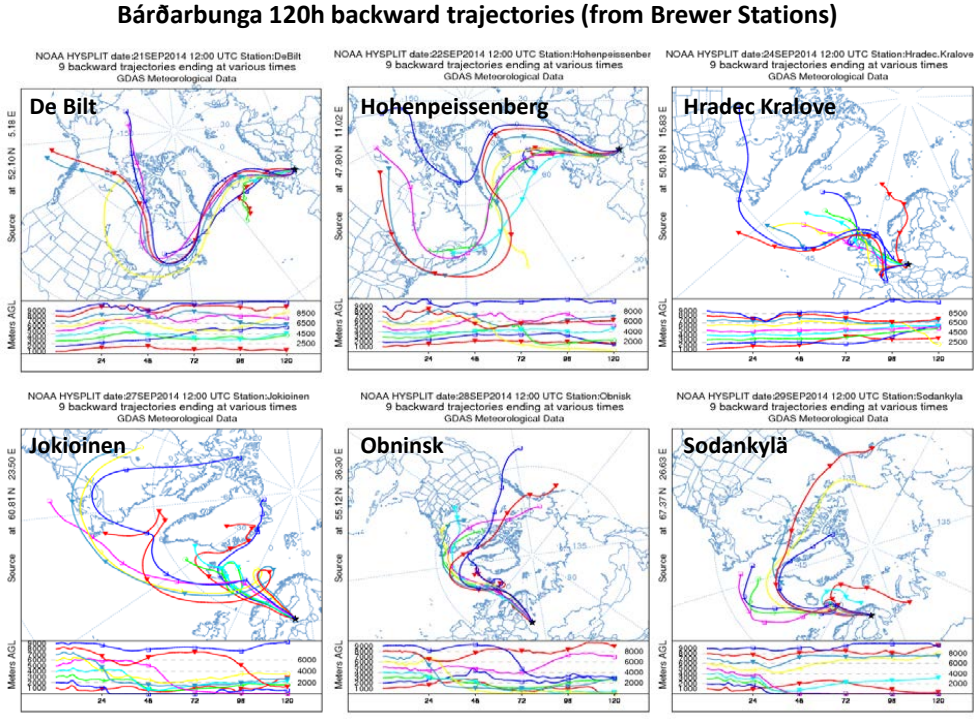

Figure 4. HYSPLIT 120 hours back trajectories of air masses arriving on the day of maximum SO_2 records for each one of the Brewer stations at De Bilt, Hohenpeissenberg, Hradec Kralove, Jokioinen, Obninsk and Sodankylä.

Atmos. Chem. Phys. Discuss., doi:10.5194/acp-2016-500, 2016
Manuscript under review for journal Atmos. Chem. Phys.
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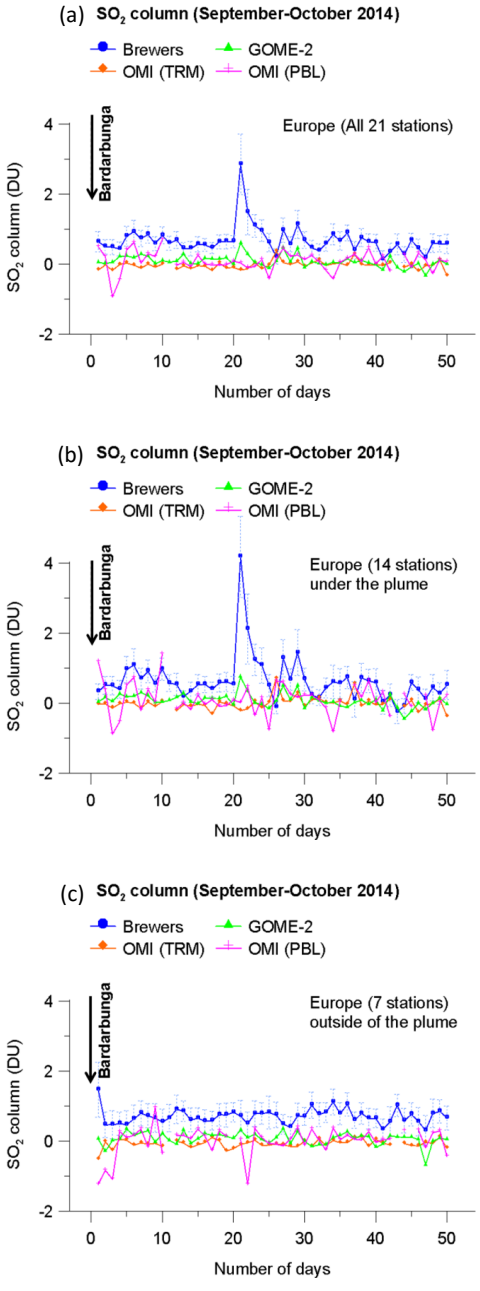

Figure 5. (a) Mean SO_2 column in DU measured by Brewers, OMI and GOME-2 during September-October 2014 over Europe. (b) Same as (a) but for stations under the plume. (c) Same as (a) but for stations outside the plume. The error bars for the Brewer observations show the standard error of all daily values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

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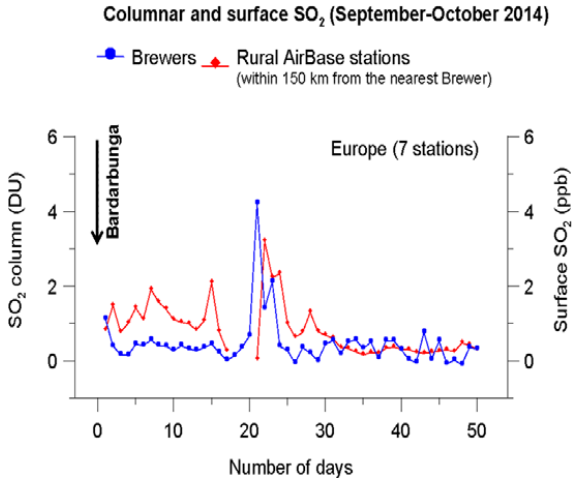

Figure 6. Mean surface SO_2 measured by Airbase class 1-2 stations located within 150 km from the nearest Brewer station.

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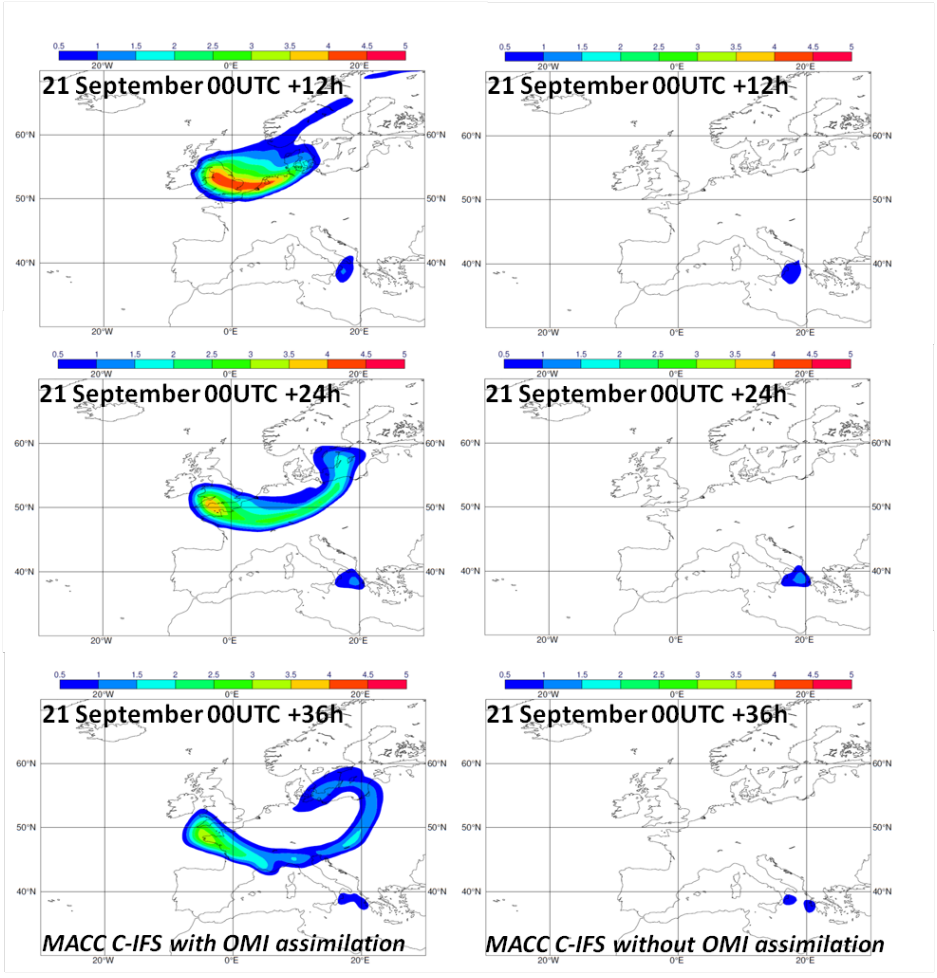

Figure 7. Charts of forecasted total column SO_2 produced within the MACC system for 21 September 2014 with OMI data assimilation (left) and without OMI data assimilation (right).

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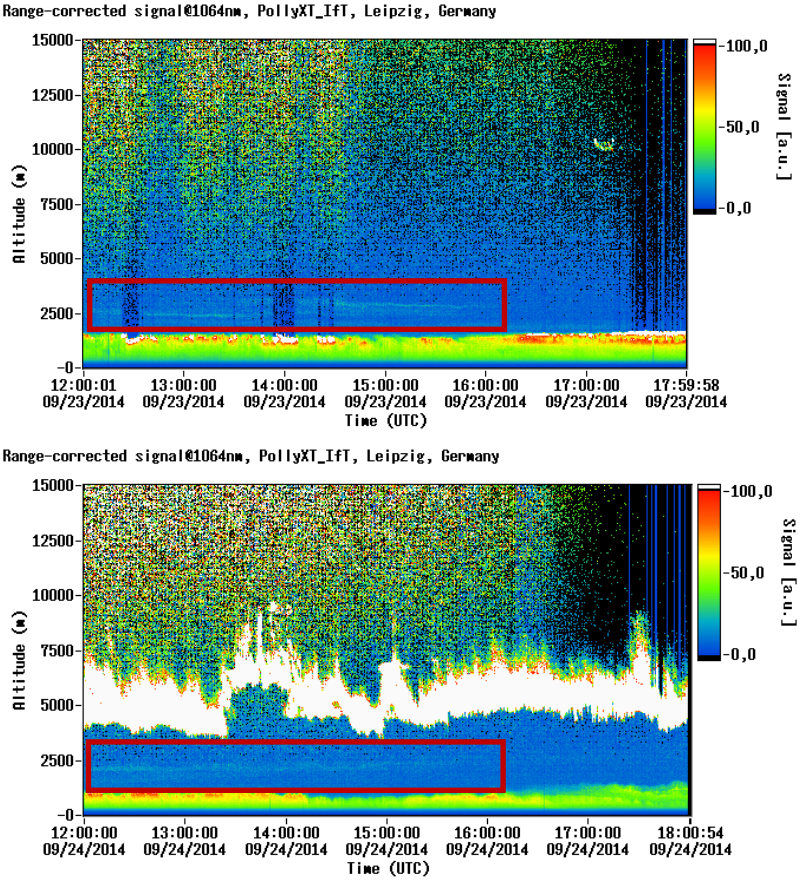

Figure 8. Range corrected signal at 1064 nm from the PollyXT lidar in Leipzig on 23 September 2014 (up) and 24 September 2014 (down). The red rectangular indicates the location of the volcanic ash layer.

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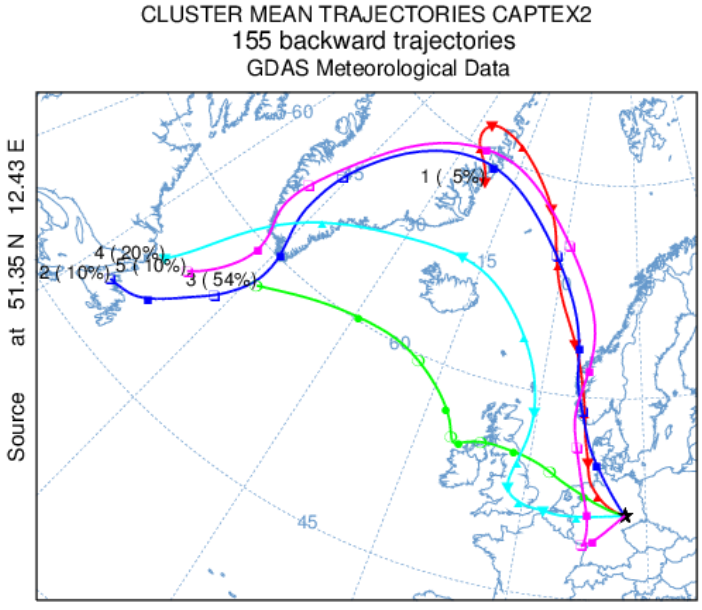

Figure 9. Cluster analysis of the HYSPLIT back trajectories that arrive every hour (from 23 September 12:00 UTC up to 24 September 18:00 UTC) at 2.5-3.5 km height over Leipzig. A 54% cluster percentage means that there is 54% chance that the SO_2 arriving anywhere between 2.5-3.5 km over Leipzig originates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

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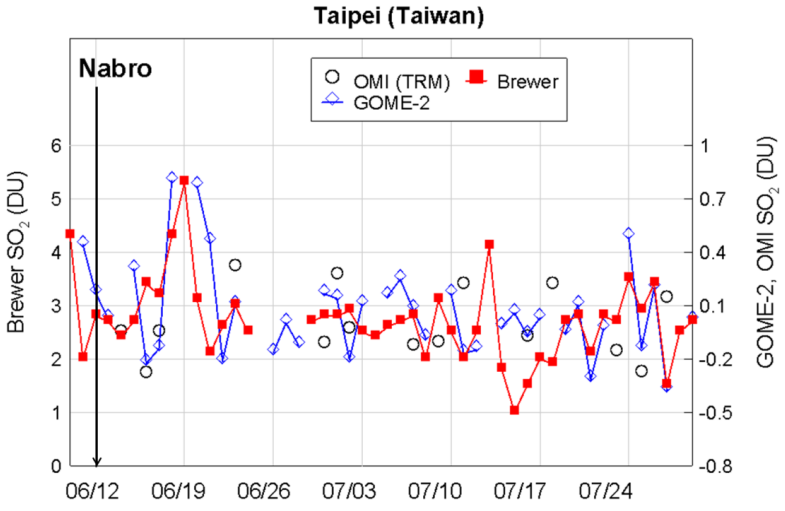

Figure 10. SO_2 columns from Brewer, OMI (TRM) and GOME-2 overpasses over Taipei, Taiwan, during June-July 2011.

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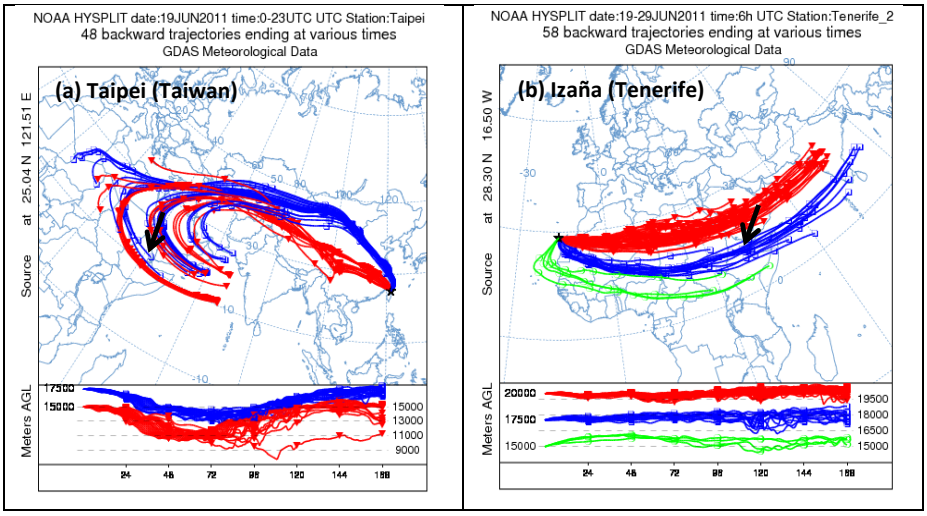

Figure 11. HYSPLIT back trajectories of air masses (a) from Taipei (Taiwan) on 19 June 2011, (b) from Izaña (Tenerife) for days 19-29 June 2011. Nabro's location is indicated by the black arrow.

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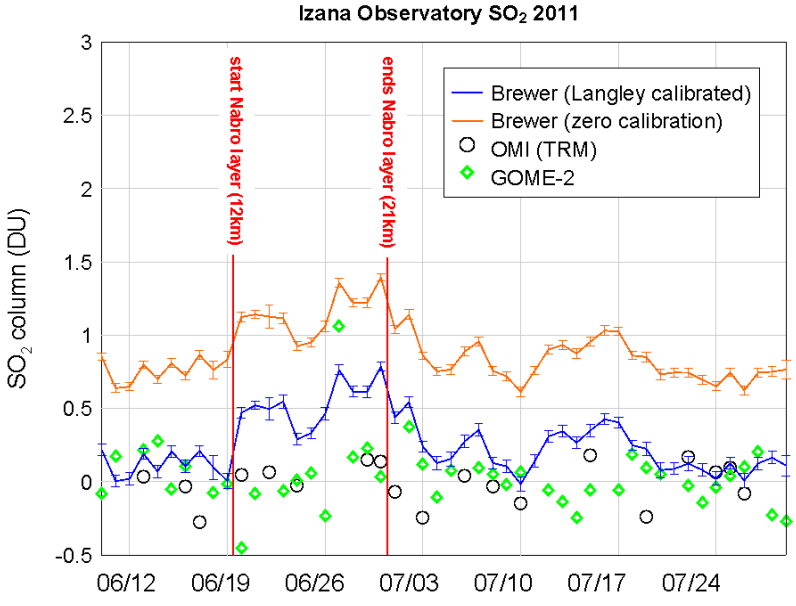

Figure 12. SO_2 calculations using the Langley calibration and the zero calibration (assuming SO_2=0 during the days 06 and 07 of June 2011). Also shown are SO_2 columns from OMI (TRM) and GOME-2 overpasses over Izaña Observatory during June-July 2011.

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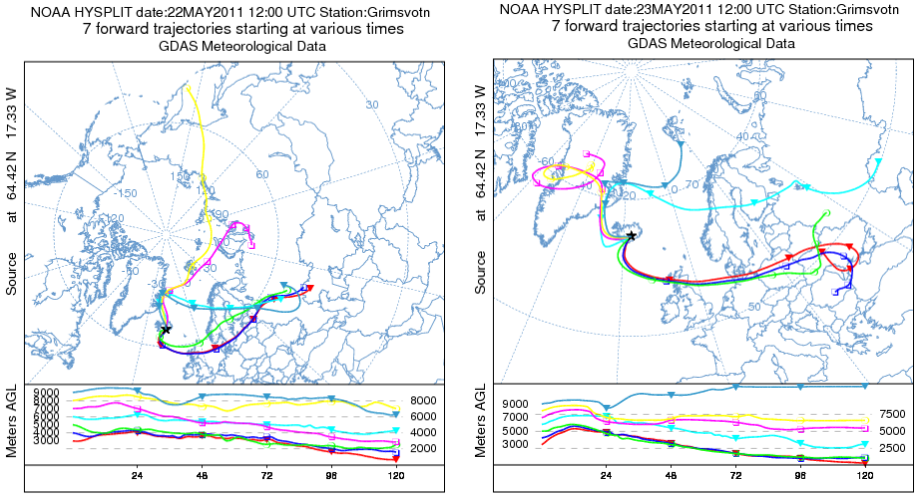

Figure 13. HYSPLIT 120 hours forward trajectories from Iceland following Grímsvötn eruption.

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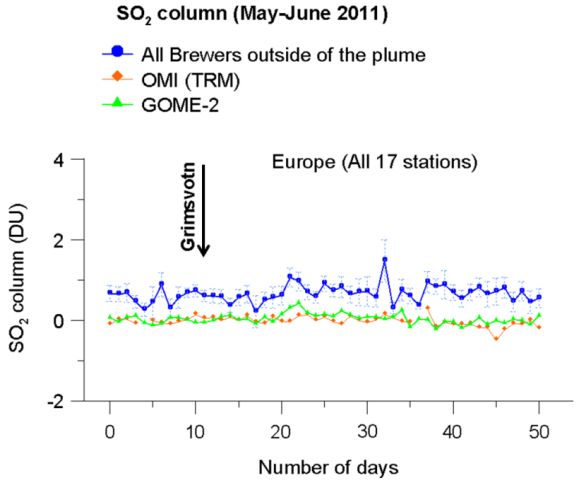

Figure 14. All Brewer stations were outside of the Grímsvötn volcanic eruption plume. The error bars for the Brewer observations show the standard error of all daily values entering the average.

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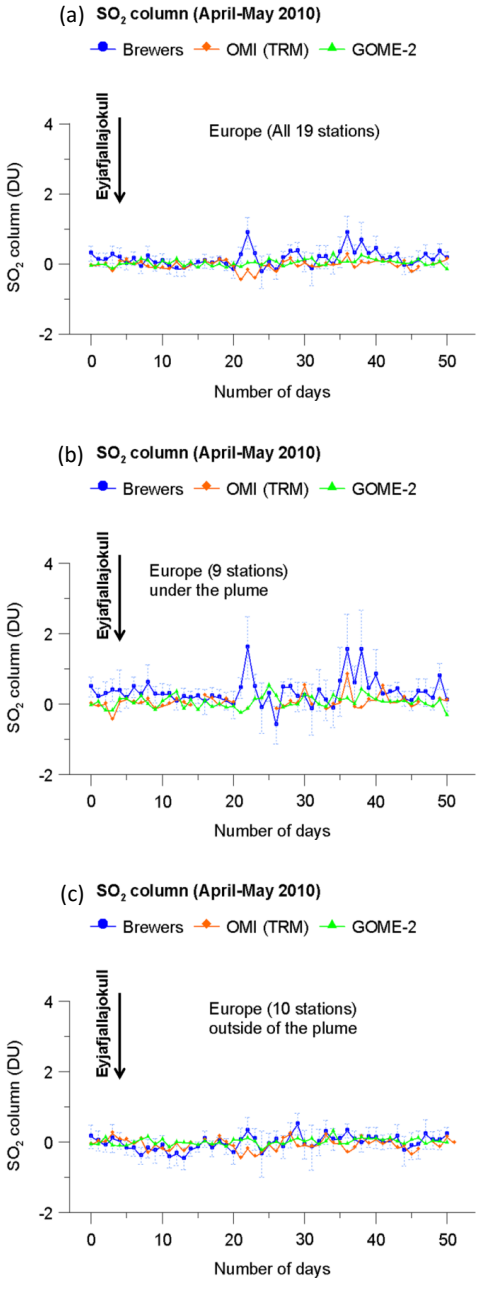

Figure 15. (a) Mean SO_2 column measured by Brewers, OMI and GOME-2 during April-May 2010. (b) Same as (a) but for stations under the plume. (c) Same as (a) but for stations outside of the plume. The error bars for the Brewer observations show the standard error of all daily values entering the average.

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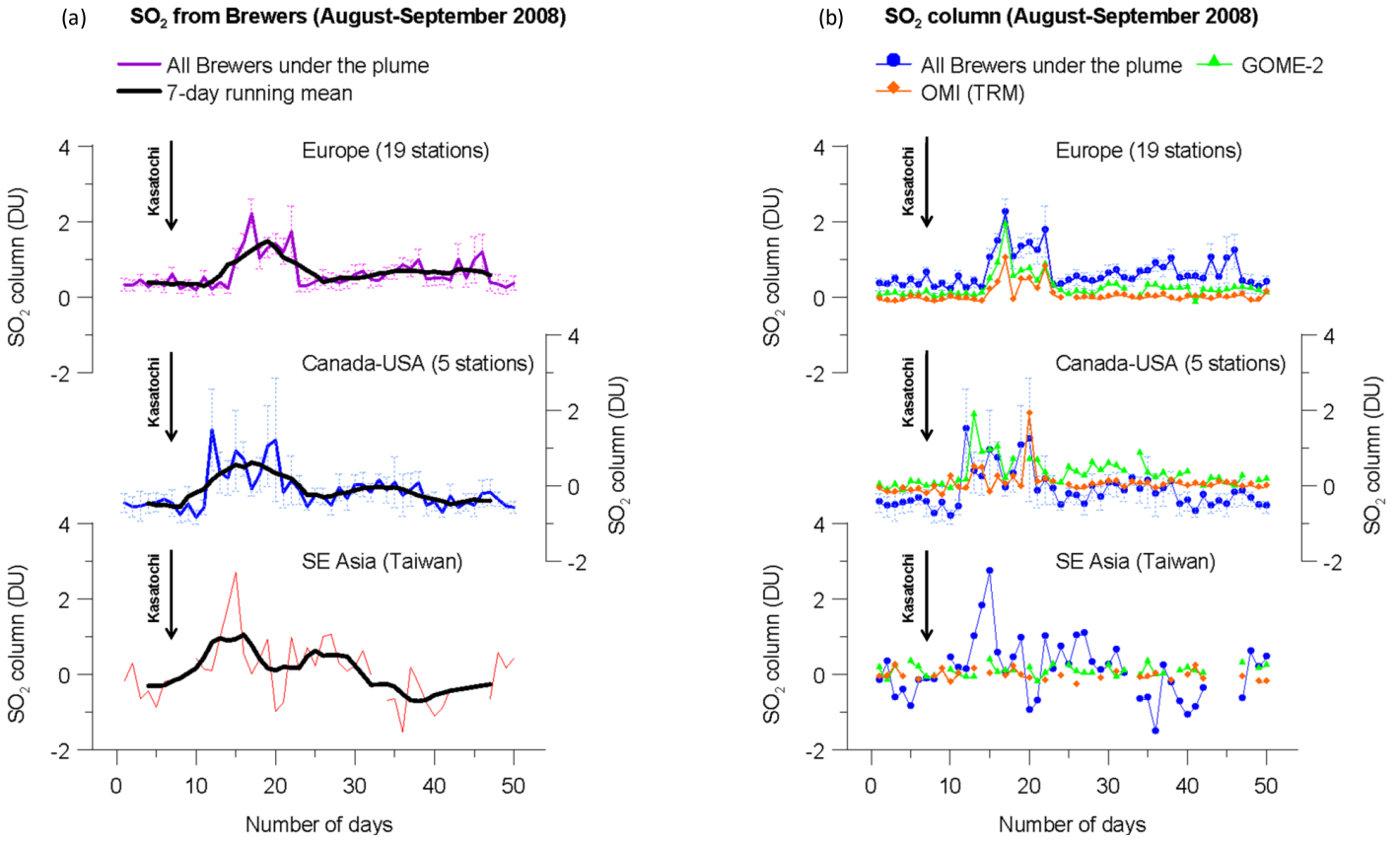

Figure 16. Mean SO_2 column measured by Brewers, OMI (TRM) and GOME-2 over Europe, Canada/USA and Taiwan during August-September 2008. The error bars for the Brewer observations show the standard error of all daily values entering the average.