

## Response to Review #1

We thank the reviewer for taking the time and appreciate the helpful comments and suggestions for improving the manuscript given in this review. We will try to change the writing style to be less descriptive and shorten the abstract.

The comments will be addressed below with review comments stated first, then the author's response in *italic*, the changes to the text is given in quotations (""), also in *italic*.

Specific comments:

Page 2, line 1: what do you mean by 'peak type' increases? Give numbers here including that date of the measurement and location.

*Included in the manuscript, changed peak type to:*

*"Surface observations in Europe showed concentration increases up to 50  $\mu\text{g}/\text{m}^3$  averaged over an hour of  $\text{SO}_2$  from volcanic plumes passing."*

You report increases in  $\text{PM}_{2.5}$  mass concentrations based on your model simulations. There are plenty of  $\text{PM}_{2.5}$  monitoring sites across Europe (many more than for  $\text{SO}_2$ ), so you ought make an effort to compare the model simulations to these observations.

*$\text{PM}_{2.5}$  observations are included in the manuscript for the station in Manchester during the first period when both  $\text{SO}_2$  and  $\text{PM}_{2.5}$  are measured at the station, for the other  $\text{PM}_{2.5}$  station with available data over the three periods the plots are in the supplementary data.*

Are there deposition measurements available that could be used to compare to the model simulations?

*When writing the manuscript before submitting to ACPD, these observations were not available. Wet deposition data are now available for some sites, and will be included in the manuscript and supplementary material.*

Page 2, line 31: state the total amount of lava produced

*Included in the manuscript*

Page 4, line 1: replace 'on the top' with 'at the top'

*Changed accordingly*

Page 4, line 4: I strongly disagree with that statement. I agree uncertainties in the source term affect both volcanic gas clouds and ash clouds, but fundamentally the processes that affect  $\text{SO}_2$  dispersion and conversion to sulfuric acid aerosol particles are different than those that affect volcanic ash concentrations downwind the source. I would simply say that Holuhraun is an eruption worth studying for gas and aerosol processes and effects.

*Removed the sentence and changed the text to:*

*"Unlike the two previous big eruptions in Iceland, Eyjafjallajökull in 2010 and Grímsvötn in 2011, this eruption did not emit ash. However, uncertainties in source estimates, time varying emissions from a point source and dependence of transport on initial injection height are similar problems for  $\text{SO}_2$  and ash plumes. For eruptions where both ash and  $\text{SO}_2$  are emitted,  $\text{SO}_2$  can act as a proxy for ash (Thomas and Prata et al, 2011; Sears et al., 2013), however separation can occur both because of*

*different eruption heights within the plume (Moxnes et al., 2014) and density differences after some time. Proven capability of modelling the transport of a volcanic plume can be useful for judging future eruption scenarios where ash may cause a problem.”*

The aims of the study could be described more clearly and put into context with previous studies (e.g. Schmidt et al., 2015, Gislason et al., 2015).

*The aim is to study the perturbed sulphur budget due to the volcanic emission, both observed and modelled. The second aim is investigate the impact of the eruption on European pollution levels. This is also made more clear in the manuscript.*

Model description:

It isn't clear to me why the Holuhraun case is called the 'control' simulation. Would it not be more intuitive to call the no\_hol simulation the control simulation?

*The control simulation is renamed basic (bas). From the observed heights, and emission fluxes given elsewhere, this simulation is the “best guess” simulation.*

You run sensitivity simulations changing the emission height, but given that your are making statements about effects on air quality, it would be better to also test the sensitivity to the SO<sub>2</sub> flux. I would recommend carrying out one simulation using 120 kt/d. It should also be possible to use a time-varying flux by using the data from Thordarson and Hartley (2015) for example.

*Increase in the SO<sub>2</sub> flux will lead to higher numbers, however the increase is close to linear to the increase in emission flux. This is also shown in the paper by Schmidt et al. (2015) and in Figure 1, where a sensitivity simulation with 120 kt/d emission (called max volc), and a simulation with the time varying Thordarson and Hartley (2015) emission is plotted (Thor volc). However comparing this simulation with the satellite data show worse result. This indicates that the height of the emission is important, and the transportation towards the station.*

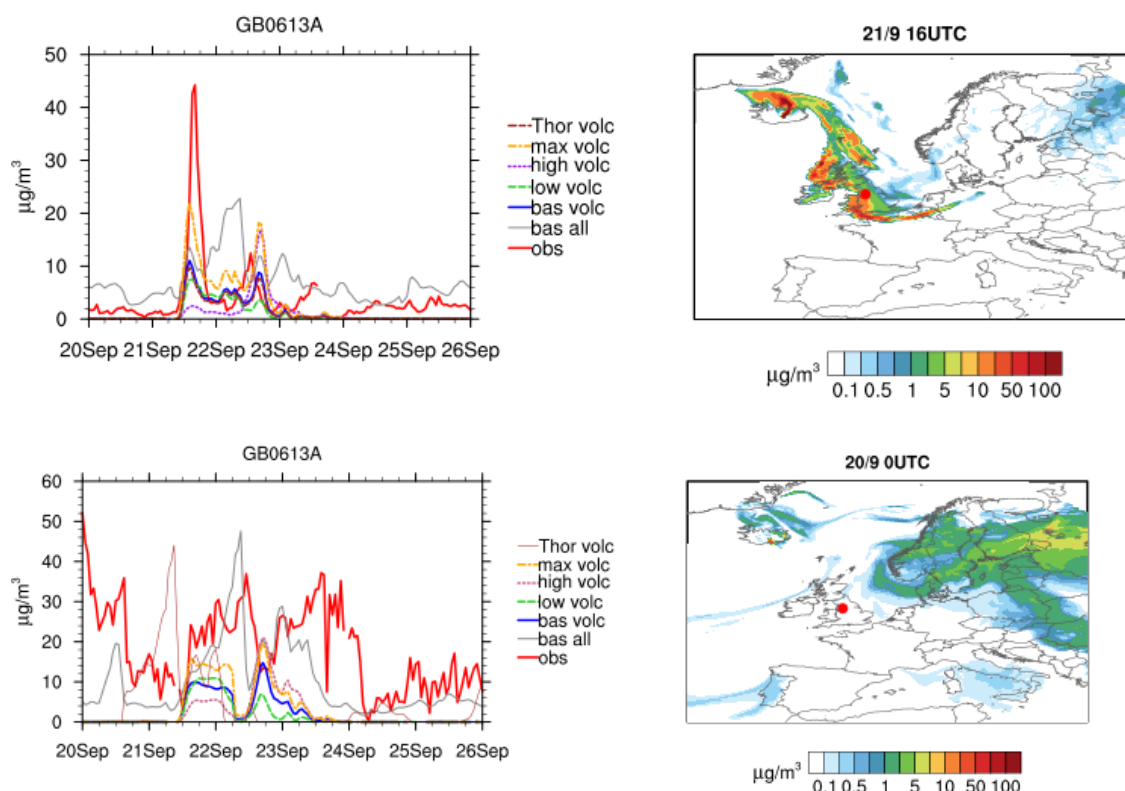


Figure 1. Measured and modelled concentration at GB0613A station in Manchester, Great Britain (red dots on the map). The timeseries above show  $\text{SO}_2$  concentrations and below for  $\text{PM}_{2.5}$  for observed (red) and five different model simulations, bas all show all sources for  $\text{SO}_2$  and  $\text{PM}_{2.5}$ , while the other volc lines only show values due to the volcanic eruption. The time of the map plot is at the maximum observed concentrations.

Observations:

Page 6, lines 23-24: Schmidt et al. (2015) used IASI to derive plume heights, which indicates that using an a priory plume profile of 7 km is too high indeed.

Changed in the manuscript to:

“As found in Schmidt et al. (2015), this is too high for the Bardarbunga eruption therefore retrieved  $\text{SO}_2$  column densities may thus be too low”

Page 8, lines 3-4: be more specific and state the dates and significance of the  $\text{SO}_2$  observations for these episodes

Included in the text:

“For the first six day period, between 20 to 26 September, high concentrations of  $\text{SO}_2$  were measured over Great Britain, and countries to the south. For the second six day period, a month later (20 to 26 October) the plume was also detected over Great Britain, but transported further east towards Germany. For the last plume studied here from 29 October to 4 November, the volcanic emission was transported southeast to the coast of Norway and countries to the south. Model data to represent the station values are picked from hourly data at model surface level in the grid cell where the station is located.”

## Results

### 3.1 Comparison to satellite data

Page 9, line 1: state the highest value for both the satellite burden and the modeled burdens.

*Added in the manuscript.*

*“The highest values are at the beginning of the period, 42.11 kt SO<sub>2</sub> for the model data on 7 September, and 37.42 kt SO<sub>2</sub> 20 September for the satellite data.”*

In particular, the simulated burdens for September 2014 should be compared to those in Schmidt et al. (2015), which should give you an opportunity to compare model performance to that of another model.

*The simulated burdens presented in this study and the simulated burdens presented in Schmidt et al. (2015) Figure 4 are not directly comparable. The model burdens are weighted with the kernel to compare to the satellite data while in Schmidt et al. (2015), the a priori satellite height in the OMI data are set to the observed heights by IASI to compare to the NAME model results with emission heights at 1.5 to 3 km. Both plots show however higher satellite burdens compared to model on 4 September and higher model burdens compared to satellite on 6 and 7 September. This is included in the discussion part of the manuscript.*

Page 10, line 7: here you should perform a sensitivity study using higher SO<sub>2</sub> emissions than 65 kt/d and discuss the comparison to the satellite-derived burdens.

*Figure 2 show the same as Fig. 2 b in the manuscript, but with the time varying emission term from Thordarson and Hartley (2015). The SO<sub>2</sub> is released in the same height as for the basic model run, between 0 and 3 km. Although matching better for the first days, the results are not better overall. All the results presented in the manuscript show that the dependency of emission height is more important. This is included in the discussion part of the manuscript.*

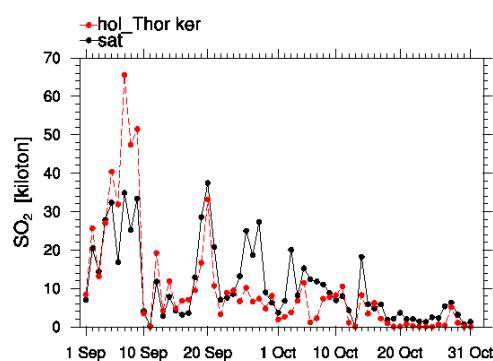
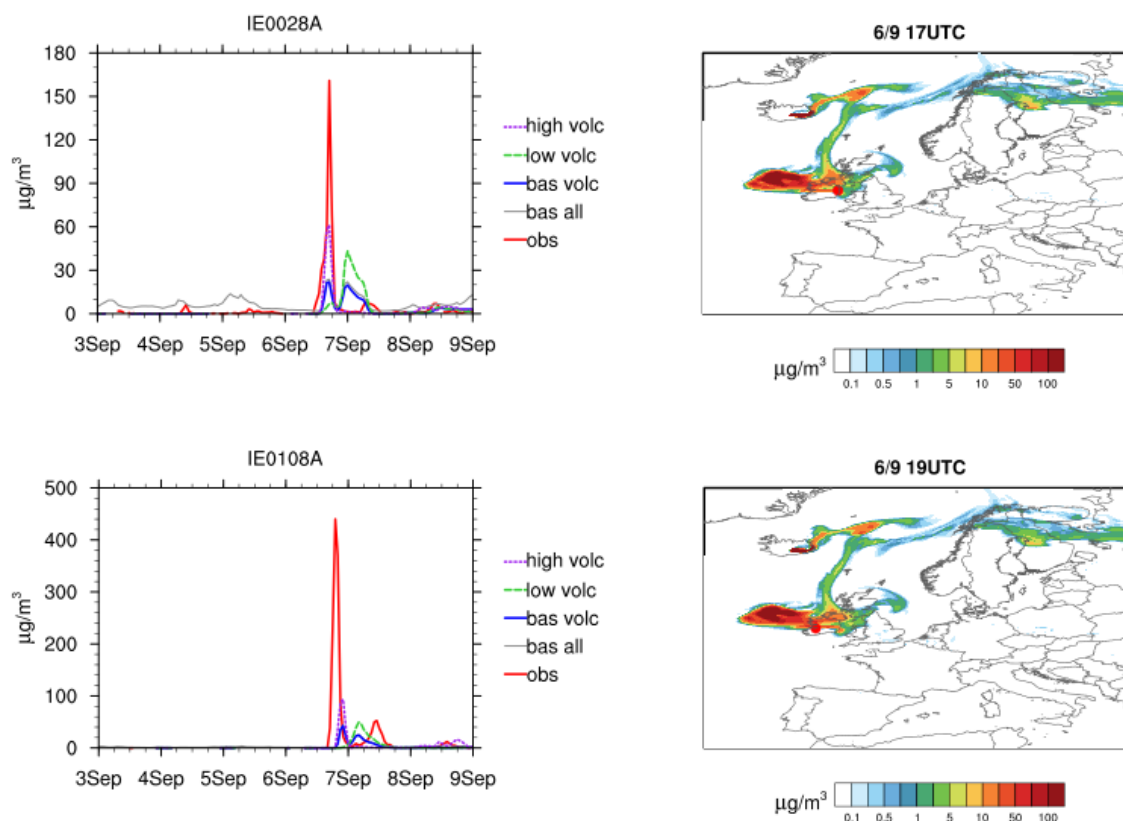


Figure 2. Daily time series of mass burdens from satellite data (black dots) and from model run with Thorarson and Hartley (2015) emission (red dots) with averaging kernel applied.

### 3.2 Surface concentrations

Page 10, line 9 onwards: give more detailed information including the locations of the measurement stations, the peak values observed and the date/time period of these observations. Surface SO<sub>2</sub> mass concentrations of about 500  $\mu\text{g}/\text{m}^3$  have been observed in Ireland on 6 September (when the eruption was most powerful). Why do you not use these data as well?

*The detailed information will be included in the manuscript. The high SO<sub>2</sub> concentration observed over Ireland on 6 September did not show up on many of the station that we were able to collect, so it was left out of the manuscript, but two Irish stations are shown in Figure 3.*



*Figure 3. Measured and modelled concentration at station IE0028A and IE0108A in Ireland (red dots on the map). The timeseries show SO<sub>2</sub> concentrations and for observed (red) and five different model simulations, bas all show all sources for SO<sub>2</sub>, while the other volc lines only show values due to the volcanic eruption. The time of the map plot is at the maximum observed concentrations.*

*Station IE0028A lies east of station IE0108A where the observed concentrations are higher. The concentration maps also show that concentrations over 100  $\mu\text{g}/\text{m}^3$  over the North Atlantic Ocean to the west of Iceland in an anticyclone. Both the stations have higher concentrations for the simulation where the emissions is put between 3 to 5 km. Schmidt et al (2015) found the same result, and analyzed the discrepancies to be a problem with the boundary layer height. The satellite comparison for this time shows that the model data have higher values than the satellite observations.*

Page 12, lines 3-4: this is only true for the later period of the eruption. You haven't analysed observational data for the early eruption phase, which should be done and it should be stated more clearly that your results support emissions of about 65 kt/d for the late Sep to Oct period.

The satellite data comparison does not clearly show that the column burdens are too low at the beginning of the period, apart from the first few days, but on September 6, the model has higher summed  $\text{SO}_2$  value than the observed satellite over the larger area (not the smaller). Both Figure 1 and Schmidt et al. (2015) found that the model runs with the higher emission altitude have higher concentrations at the sites, the satellite time series of this model simulations show that the model data have even higher values (Figure 4). These results points in two different directions, and it is difficult to conclude that the emission flux should be higher and at a higher level although Schmidt et al. (2015) found this. The higher concentrations in the observations seem to come from the boundary layer being badly represented in the meteorological data. This is included in the discussion part of the manuscript.

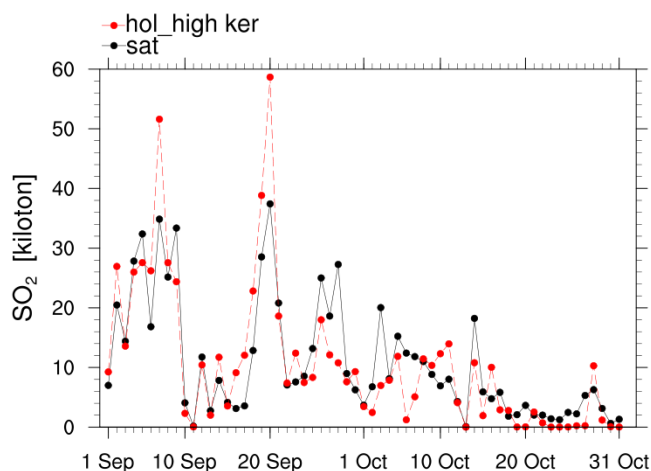


Figure 4. Daily time series of mass burdens from satellite data (black dots) and from model run with emissions released between three and five km, high\_hol (red dots) with averaging kernel applied.

To maybe clarify more, the text is changed to:

“Overall the comparison to observations, both satellite and station data, the bas\_hol model simulation match best with the observed satellite column burdens and with the timing and for some stations concentrations of the observed peaks.”

### 3.3 Effects of the eruption on European pollution

Page 12, lines 6-7: this has also been shown by Gislason et al. (2015) and Schmidt et al. (2015)

Added the references.

Page 12, line 18 onwards: rewrite all paragraphs using less descriptive writing style

Will change the writing style

The increases in simulated  $\text{PM}_{2.5}$  mass concentrations ought to be compared to measurements from across Europe otherwise the discussion is of little scientific value (in particular because the model is not capturing peak  $\text{SO}_2$  mass concentrations at the ground compared to the observations).

$\text{PM}_{2.5}$  is included in the station comparisons, where a station both measures  $\text{PM}_{2.5}$  and  $\text{SO}_2$  in the paper and the other stations in supplementary material.

## 4 Discussion

First paragraph: several aspects of this discussion are too simplistic because there are observations of the plume height (both at the source and in the far-field using IASI for example)

*Although presenting plume heights, Schmidt et al. (2015) does not use these heights for their model simulations, and there are some discrepancies in the calculations especially the am data on 15 September where the center of mass is 4 km and the plume height is only 3.9 km. The authors agree that the height is not unknown so included it in the discussion.*

*Changed the text to:*

*“d) Schmidt et al. (2015) presents IASI (Infrared Atmospheric Sounding Interferometer) plume heights between 5.5 km to 1.6 km derived from an area of 500 km around the volcanic location, and a mean IASI centre of mass height between 2.7 km to 0.6 km. The fluctuating real height of the SO<sub>2</sub> plume may introduce additional bias between model and satellite VCDs.”*

Second paragraph: Unless you carry out a sensitivity study changing the SO<sub>2</sub> flux, you must not state that the variations in the source flux explain the differences between the observations and your model results because you haven't demonstrated that.

*Model runs with different emission fluxes are presented in the answer here. The almost linear increase of concentrations with emission is also presented in Schmidt et al. (2015). Variations in emission flux can also change within an hour, so unless a more thoroughly study is done for the emission term, this is an uncertainty factor.*

Page 15, lines 16-26: state the date and station name for each event that you discuss. I struggle to understand why the difference between the modeled and observed concentrations for the 6 Sep 2014 air pollution event cannot be explained by higher emissions fluxes.

*Added the information in the text.*

*For the 6 September event, as discussed above, the satellite results and the concentrations at the stations show discrepancies in terms of concentrations, other studies points towards a higher emission during this first week. The models (both EMEP and NAME) fail to simulate the high concentrations even with higher emissions. Schmidt et al. (2015) points towards the model not being able to reproduce the atmospheric subsidence and the representation of the boundary layer from the meteorological field.*

## Conclusions

All paragraphs need to be rewritten in a less descriptive manner.

*Will change the writing style*

Page 16, line 20: 'increase in SO<sub>2</sub>' what? Is there a word missing? Do you mean burden or surface mass concentrations? Previous studies that came to the same conclusion should be referenced here.

Changed the sentence to:

*The increase in emitted SO<sub>2</sub> to the atmosphere caused by the volcanic eruption at Holuhraun were observed by satellite and detected at several stations over Europe (Schmidt et al. 2015).*

Last sentence: I disagree; the increase in SO<sub>2</sub> mass concentrations was significant in several places even though the pollution episodes were transient.

*Changed it to:*

*“Even with high emissions from the volcanic fissure at Holuhraun, the increase in pollution levels over Europe is low, with only transient episodes with high increases in SO<sub>2</sub> concentration.”*

Figure 1: state which model run is shown.

*Added to the caption.*

Figure 3: give date range and how does this compare to Schmidt et al. (2015) who I presume used the same satellite data but state much higher burdens than reported here. Is this down to different averaging periods?

*This is explained above. The a priori height used by the retrieval of OMI satellite is different.*

*References:*

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. (2015) Environmental pressure from the 2014–15 eruption of Bárðarbunga volcano, Iceland. *Geochem. Persp. Let.* 1, 84-93.

Schmidt, A., S. Leadbetter, N. Theys, E. Carboni, C. S. Witham, J. A. Stevenson, C. E. Birch, T. Thordarson, S. Turnock, S. Barsotti, et al. (2015), 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.

Thordarson, T. & Hartley, M. (2015): Atmospheric sulfur loading by the ongoing Nornahraun eruption, North Iceland. , 17, 10708.



## Response to Review #2

We thank the reviewer for taking the time and appreciate the helpful comments and suggestions for improving the manuscript given in this review.

The comments will be addressed below with review comments stated first, then the author's response in *italics*, the changes to the text is given in quotations (""), also in *italics*.

**Structure and title:** although the title of the paper focuses on the air pollution effects of the Holuhraun fissure eruption, the text is unbalanced in this regard, with a lot of description on the comparison of EMEP simulations with satellite and ground-based measurements. The title should be changed accordingly or the text restructured and reduced. A potential title, matching better the content of the paper, could be "A model study of the three months of the Holuhraun volcanic fissure: comparison with satellite and ground-based data and air pollution effects". The same unbalance exists in the, too long (please reduce), abstract. If the title remains the same, then the paper structure should be modified and the sections on the comparison with ground-based and satellite data should be gathered into a specific section that addresses the performance of the model calculations for this event. The results and discussion should then focus solely on the air pollution aspects once the following item is also addressed.

*The abstract will be shortened, and the authors agree that the title does not reflect the context of the manuscript, title is changed to:*

*"A model study of the pollution effects of the first three months of the Holuhraun volcanic fissure: comparison with observations and air pollution effects"*

**Air pollution effects and chemical transport model results:** the results and discussion on the air pollution effects should be further extended. The text is based solely on one model simulation with evident limitations. More discussion should appear on the potential effects of the mentioned limitations in the overall air quality side of the paper. In addition, the authors present wet and dry deposition results of the simulations with no comparison with existing data. Whenever wet scavenging data exists for such episode, it should be used to assess the very important effect of scavenging. The chemical transport model results are presented and discussed but without the required depth: why are there such large differences in the modelling results and the measurements? Is there a problem in the atmospheric mixing of the EMEP model that leads to such poor representation of the ground base measurements? what are the potential causes of not only the magnitude differences of the modelled versus measured peaks but also in their times? Have they tested different meteorological fields? Although it is clear that a thorough analysis would probably be out of the scope of the paper, additional thought should be made and added to the manuscript to help the reader with the questions that will surely appear when looking at Figures 4 to 6.

*More on the limitations of the model for not performing better for the high concentration events will be included in the discussion part. Comparison to  $PM_{2.5}$  measurements and  $SO_x$  wet deposition measurements will be included for stations where it is available. There is no known problem in the atmospheric mixing in the EMEP model. The complex transport to the stations for the first episode with first southerly winds, then northerly caused the  $SO_2$  to stay in the atmosphere longer and increase in concentrations. The uncertainties due to model representations and meteorology errors accumulate and create the discrepancies seen in Figure 4. The comparison is better for the two later periods. The ECMWF meteorology is the best available meteorology for the EMEP model, and the resolution is also high. Schmidt et al. (2015) use another meteorological driver and also find the same*

*discrepancies over this late September period. The result and discussion part will be extended to include more station comparison data.*

#### Specific comments

Abstract: the abstract is too long and unfocused. Please highlight the main results according to the title of the paper (see General Comments)

*The abstract will be shortened and more focused.*

Abstract Line 12 - “lava floated” I would change float by flow.

*Changed accordingly.*

Line 4 Pag. 4 - The authors stated that this case can be used as a proxy for ash events as well. As the authors state further on (lines 9-10) that might not be the case, as Grimvoetn event showed with significantly different transport patterns for SO<sub>2</sub> and ash. In addition the processes occurring for ash (including fine and coarse ash, aggregation, gravitational settling...) and SO<sub>2</sub> (gas and aqueous phase chemistry) are different enough to add different uncertainties into the processes. It is indeed true that uncertainties in the source term may dominate, but I would rather suggest the authors erase the sentence “The Holuhraun eruption can also serve as a prototype...”

*The authors agree that SO<sub>2</sub> is not a prototype for ash, removed the statement and changed the text to:*

*“Unlike the two previous big eruptions in Iceland, Eyjafjallajökull in 2010 and Grímsvötn in 2011, this eruption did not emit ash. However, uncertainties in source estimates, time varying emissions from a point source and dependence of transport on initial injection height are similar problems for SO<sub>2</sub> and ash plumes. For eruptions where both ash and SO<sub>2</sub> are emitted, SO<sub>2</sub> can act as a proxy for ash (Thomas and Prata et al, 2011; Sears et al., 2013), however separation can occur both because of different eruption heights within the plume (Moxnes et al., 2014) and density differences after some time. Proven capability of modelling the transport of a volcanic plume can be useful for judging future eruption scenarios where ash may cause a problem.”*

Section 2.1 Model description: it would be useful to the reader to have more information on how the chemical module of EMEP/MSC-W works for SO<sub>2</sub> since for this event the reactions with both OH and in the aqueous-phase (due to its low altitude pathway towards Europe) are significant.

*Extended the model description to:*

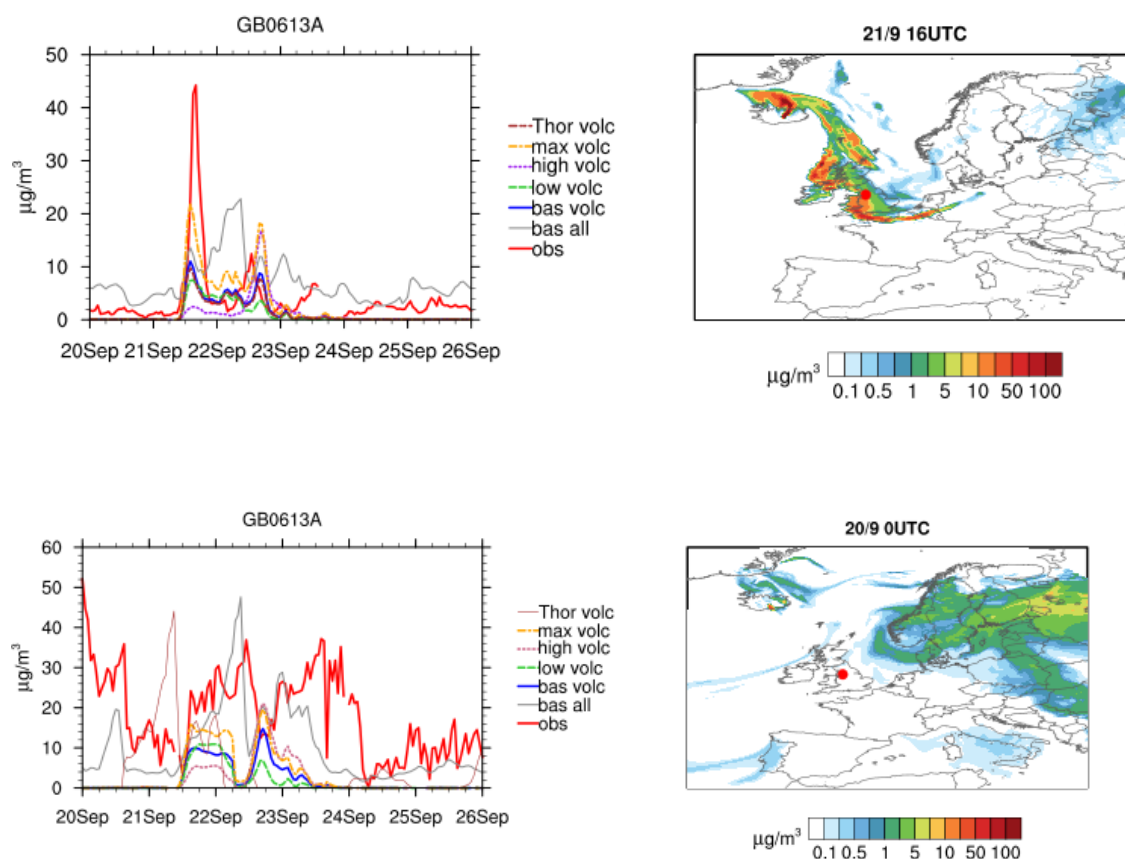
*“SO<sub>2</sub> is oxidized to sulphate in both gas and aqueous phase with assumed equilibrium. In gas phase the oxidation is initiated by Hydroxide (OH), OH is labelled “short lived” and is controlled by local chemistry. In aqueous phase the oxidants ozone, hydrogen peroxide and oxygen catalysed by metal ions contribute to oxidation.”*

Line 12 Pag. 4 - The authors should rewrite this paragraph in order to make it clearer to the reader what are they actually aiming at. What is the MAIN aim? and to achieve such aim what are the SECONDARY milestones or aspects that are addressed?

*The aim is to study the perturbed sulphur budget due to the volcanic emission, both observed and modelled. The second aim is investigate the impact of the eruption on European pollution levels. This is also made more clear in the manuscript.*

Line 16 Pag. 5 - Can the authors state (and even better reference) why they are finally using a constant 750 kg/s SO<sub>2</sub> flux? They could have easily implemented a variable emission or taken a “worst case scenario” with the maximum flux of 120kt/day. This affects the discussion on the air pollution section and therefore should be clarified and its implications on the air quality results clearly discussed.

*A worst case scenario with a emission of 1400 kg/s (max\_hol) and a time varying emission given in Thordarson and Hartley (2015)(Thor\_hol) is also studied, but the results were not better compared to observations. As shown in the Figure , Ifor concentration comparison at the Manchester station in September where it is shown that an increase in emission gives an almost linear increase in concentrations of SO<sub>2</sub> and PM<sub>2.5</sub> (and deposition, not shown). Figure 2 show the satellite comparison for the hol\_Thor simulation, same as Figure 2b in the manuscript.*



*Figure 1. measured and modelled concentration at GB0613A station in Manchester, Great Britain (red dots on the map). The timeseries above show SO<sub>2</sub> concentrations and below for PM<sub>2.5</sub> for observed (red) and five different model simulations, bas all show all sources for SO<sub>2</sub> and PM<sub>2.5</sub>, while the other volc lines only show values due to the volcanic eruption. The time of the map plot is the time of maximum observed concentration.*

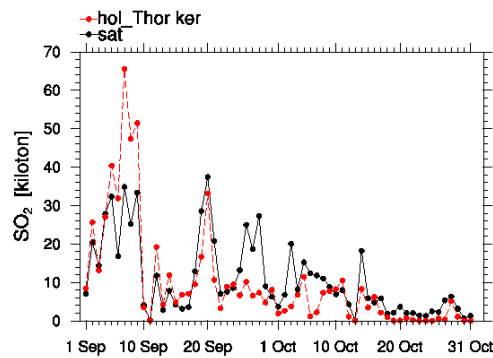


Figure 2. Daily time series of mass burdens from satellite data (black dots) and from model run with Thorarson and Hartley (2015) emission (red dots) with averaging kernel applied.

The height of the emission is seen to be more important, and therefore these two simulations are not included in the manuscript. This discussion is included in the manuscript, and the number behind the emission is added in the text:

“Emission from the Holuhraun fissure is set to a constant 750 kg/s  $SO_2$  (65 kt/d) for the entire simulation from the total  $2.0 \pm 0.6$  Tg  $SO_2$  emitted in September estimated in Schmidt et al. (2015). “

Line 21-23 Pag. 5 - if the authors explain what the control run consists of, also the low and high runs should be explained in addition to the reference of table 1.

Will include more description.

Line 4 Pag. 8 - The measurements were regridded? following what method?

The sentence is changed to:

“Model data to represent the station values are picked from hourly data at model surface level in the gridpoint where the station is located.”

Line 10-12 Pag. 12 - It is not entirely clear how the gross numbers in Table 2 are obtained. Is it for the 31 countries but the text states “only grid cells covering ONE ...”.

Thank you for pointing out that this it is not clear. The sentence is changed to:

“Grid cells covered by the countries mentioned are used for calculating the results shown in the table,”

Section 3.3 “Effects of the eruption on European pollution”. As stated in the general comments, this section should be extended. In addition, the authors should be careful with too general statements when their conclusions are based solely in one small set of simulations which, from the previous sections, do not prove to be very representative of the concentrations at ground level. Also, please try to add comparisons, whenever possible, with wet deposition measurement data.

The section will be extended to include more comparison to the station data observations, and rewritten so the statements better reflect the uncertainty that comes from a single model study.

## References:

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. (2015) Environmental pressure from the 2014–15 eruption of Bárðarbunga volcano, Iceland. *Geochem. Persp. Let.* 1, 84-93.

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Thordarson, T. & Hartley, M. (2015): Atmospheric sulfur loading by the ongoing Nornahraun eruption, North Iceland. , 17, 10708.

# A model study of the pollution effects of the first three months of the Holuhraun volcanic fissure: comparison with observations and air pollution effects

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## Abstract

The volcanic fissure at Holuhraun, Iceland started at the end of August 2014 and continued for six months to the end of February 2015. Lava ~~floated~~flow onto the Holuhraun plain ~~associated~~combined with ~~large~~-SO<sub>2</sub> emissions amounting up to approximately 4.5 times the daily anthropogenic SO<sub>2</sub> emitted from the 28 European Union countries, Norway, Switzerland and Iceland. In this paper we present results from EMEP/MSC-W model simulations where we added 750 kg/s SO<sub>2</sub> emissions at the Holuhraun plain from September to November. ~~The emission amounted to approximately 4.5 times the daily anthropogenic SO<sub>2</sub> emitted from the 28 European Union countries, Norway, Switzerland and Iceland. at three different emission heights.~~ Model results are compared to satellite observations and European surface measurements. ~~The dispersion but also the ambiguity of the satellite data, due to what is assumed in the retrieval as a priori SO<sub>2</sub> profile, is further explored with model sensitivity runs using different emission height distributions from the volcano. Satellite-comparable adjusted model vertical column densities are calculated for the~~The different sensitivity runs where the SO<sub>2</sub> mixing ratios from different vertical layers are runs are weighted with the satellite averaging kernel. ~~The results show the importance of using the averaging kernel when comparing the model to satellite column loads, the maximum column densities over 10 DU in the original model data are reduced by around 50 % due to the , the effect of the~~ weighting. ~~For most days the satellite retrievals have higher mass burdens values than the adjusted model~~

when summed up over the North Atlantic area. The discrepancies are explained by the unrealistic constant emission term in the model simulations, and because the area used for the summation is are dependent on the satellite data detection limit, and the correct position of the model SO<sub>2</sub> plume height of the sulphur dioxide in the atmosphere. Surface observations in Europe showed peak-type concentration increases up to 50 µg/m<sup>3</sup> averaged over an hour of SO<sub>2</sub> ~~concentrations~~ from volcanic plumes passing ~~by and lasting only for a short time.~~ Three well identified episodes are documented for more in detail. For all the events the timing of the observed concentration peaks compared ~~to the model~~ quite well. ~~For the first episode presented, the model concentrations are only about 10% to 40% of the observed concentrations. The transport of SO<sub>2</sub> to Europe during this event is found to contribute to very high measured and modelled concentrations at the stations. For the later plumes, the observed and model concentrations at the stations compare better in magnitude.~~ to the model results.

The overall changes in the European SO<sub>2</sub> budget due to the volcanic fissure are estimated. ~~SO<sub>x</sub> three~~ Three monthly wet deposition of SO<sub>x</sub> in the 28 European Union countries, Norway and Switzerland is found to be more than 30 % higher in the ~~control~~ model simulation with Holuhraun emission compared to a model simulation with no Holuhraun emission. The ~~biggest~~ largest increases, apart from extreme values on Iceland, are found on the coast of Northern Norway, a region with frequent precipitation during westerly winds. ~~The total deposition levels in this region become equal to the most polluted regions over Europe and the average model deposition for Norway is doubled the level it was back in 1990.~~

~~For~~ Average SO<sub>2</sub> and PM<sub>2.5</sub> surface concentrations, ~~there is increase by~~ only ~~a~~ ten and six percent ~~increase~~ over Europe ~~between the two model simulations~~, respectively. Although the percent increase of PM<sub>2.5</sub> concentration is highest over Scandinavia and Scotland, an increase in PM exceedance days is found over Ireland and the ~~Benelux region. Especially the Benelux region is~~ already ~~very~~ polluted, ~~so that~~ Benelux region, where a small increase in pollution leads to an increase in exceedances days. ~~Although there was a large increase in total daily emission of SO<sub>2</sub> over Europe due to the eruption, Iceland is located too far away to make a large impact on average pollution levels in the European countries, except in Iceland itself.~~

## 1 Introduction

Increased seismic activity in the Bárðarbunga volcano was recorded by the Icelandic Met Office from the middle of August 2014 (<http://en.vedur.is/earthquakes-and->

volcanism/volcanic-eruptions/holuhraun/). The activity continued in the volcano but some tremors appeared also towards the Holuhraun plain, a large lava field north of the Vatnajökull ice cap, the latter covering the Bárðarbunga and Grimsvötn volcano. On August 31 a continuous eruption started at Holuhraun with large amounts of lava pouring onto the plain and large amounts of sulphur dioxide (SO<sub>2</sub>) emitted into the atmosphere (Sigmundsson et al. 2015). Thordarson and Hartley (2015) estimated SO<sub>2</sub> emissions from the magma at Holuhraun to be around 30 kt/d to 120 kt/d over the first three months of the eruption, with a maximum during the first two weeks of September. Schmidt et al. (2015) also found that among several model simulations with different emission fluxes, the model simulations with the largest emission (120 kt/d) compared best with satellite observations at the beginning of September. In comparison, Kuenen et al. (2009) estimated the daily anthropogenic emission from the 28 European Union countries for 2009 to be 13.9 kt/d, while the 2013 estimate is 9.8 kt/d (EMEP, 2015). The eruption ended in February 2015 and during the 6 months of eruption a total of approximately 11 (± 5) Tg SO<sub>2</sub> may have been released (Gislason et al. ~~2015~~-2015), and the total lava field from the fissure were 85 km<sup>2</sup> with a volume of 1.4 km<sup>3</sup> (vedur.is). It is of interest to investigate the impact of these volcanic emissions on current SO<sub>2</sub> levels in Europe. In the last decades, measures have been taken to reduce SO<sub>2</sub> emissions, triggered by the Convention on Long-range Transboundary Air Pollution (LRTAP), in Europe. Significant reductions of 75% in emission between 1980 and 2010 are confirmed by observations (Torseth et al., 2012). The impact of volcanic eruptions with SO<sub>2</sub> emissions can thus perturb the European atmospheric sulphur budget to a larger extent than before and potentially lead to new acidification of lakes and soils if the eruption would last over a long time period.

For comparison, the big 1783 Icelandic Laki eruption lasted eight months and released a total amount of estimated 120 Tg of SO<sub>2</sub> ~~over eight months~~. The resulting sulphuric acid caused a haze observed in many countries of the northern hemisphere and increased mortality in Northern Europe (Grattan et al., 2003, Thordarson and Self, 2003, Schmidt et al., 2011). The fissure at Holuhraun was much weaker than the Laki fissure, both in terms of amount of SO<sub>2</sub> released and probably also the height of the eruptive column. Thordarson and Self (1993) estimated that the Laki erupted at emission heights up to 15 km, while the observations of the Holuhraun eruptive cloud saw the plume rising up to 4.5 km (vedur.is). Ground level concentrations exceeded the Icelandic hourly average health limit of 350 µg/m<sup>3</sup> over large parts of Iceland (Gislason et al. 2015). The World Health Organization (WHO) has a 10 minute limit of 500 µg/m<sup>3</sup> and a 24-hour limit of 20 µg/m<sup>3</sup>. High hourly mean surface



1 concentrations of SO<sub>2</sub> were measured in Ireland (524.2 µg/m<sup>3</sup>), but then also in Austria (247.0  
2 µg/m<sup>3</sup>) and Finland (180 µg/m<sup>3</sup>) (Schmidt et al. 2015, Ialango et al. 2015).

3 A climate impact of high SO<sub>2</sub> emissions may be suspected, such as a cooling of climate due to  
4 an increase in aerosol loadings. Gettelman et al. (2015) using a global climate model found a  
5 small increase in cloud albedo due to the Holuhraun emissions resulting in -0.21 Wm<sup>-2</sup>  
6 difference in radiative flux ~~on~~at the top of the atmosphere. If the event had happened earlier  
7 in the summer a larger radiative effect could be expected (-7.4 Wm<sup>-2</sup>). Understanding the  
8 atmospheric sulphur budget associated to such events is thus of great interest also for climate  
9 science. ~~The Holuhraun eruption can also serve as a prototype to study ash transport from an~~  
10 ~~Icelandic volcano.~~ Unlike the two previous big eruptions in Iceland, Eyjafjallajökull in 2010  
11 and Grímsvötn in 2011, this eruption did not emit important amounts of ash ~~that disrupted air~~  
12 ~~traffic.~~ However, uncertainties in volcanic source estimates, time varying emissions from a  
13 volcano type of point source, and dependence of transport on initial injection height, ~~transport~~  
14 ~~and removal processes from Iceland to Europe~~ are similar problems for SO<sub>2</sub> and ash plumes.  
15 ~~Despite Moxnes et al. (2014) showing that~~ For eruptions where both ash and SO<sub>2</sub> and ash are  
16 emitted, SO<sub>2</sub> can have ~~acted~~ as a proxy for ash (Thomas and Prata et al, 2011; Sears et al., 2013),  
17 however separation will occur because of density differences and different eruption heights;  
18 ~~proven~~ (Moxnes et al., 2014). Proven capability of modelling the transport of a SO<sub>2</sub> volcanic  
19 plume can be useful for judging future eruption scenarios where SO<sub>2</sub> or ash can cause a  
20 problem.

21 ~~This study will~~ The Holuhraun eruption is worth being analysed for several gas and aerosol  
22 transport and transformation processes, this study will mainly focus on simulated air quality  
23 effects and the perturbed sulphur budget due to the volcanic SO<sub>2</sub> emissions during the first  
24 three months, ~~the first two covered also by satellite observations. of the eruption.~~ Several  
25 stations in Europe reported high concentrations of SO<sub>2</sub> during this time and case studies are  
26 chosen to evaluate simulated plume development over Europe. The transport is modelled with  
27 the EMEP/MSC-W chemical transport model, one of the important models used for air  
28 quality policy support in Europe during the last 30 years (Simpson et al. 2012). The first two  
29 months of the eruption are well covered by satellite observations. Both station and satellite  
30 data are compared to model results to understand the amplitude and magnitude of the sulphur  
31 budget perturbation. ~~A big uncertainty for any volcanic eruption modelling is the emission~~  
32 ~~term, both with respect to height and magnitude~~ The effect of the plume. ~~The impact of the~~

~~height distribution of the emissions~~injection height on the model results is studied ~~throughby~~ sensitivity simulations. Finally the perturbed European sulphur budget, ~~as simulated by the EMEP/MSW model,~~ is documented and discussed to investigate the impact of increased SO<sub>2</sub> emission from a Icelandic volcano on European pollution levels.

## 2 Methods

### 2.1 Model description

The model simulations of the transport of the SO<sub>2</sub> Holuhraun emissions are done with the 3-D Eulerian chemical transport model developed at the Meteorological Synthesizing Centre-West (MSC-W) for the European Monitoring and Evaluation Programme (EMEP). The EMEP/MSW model is described in Simpson et al. (2012). ~~Sulphate production from SO<sub>2</sub> in both gas phase and aqueous phase are accounted for.~~SO<sub>2</sub> is oxidized to sulphate in both gas and aqueous phase. In gas phase the oxidation is initiated by OH and is controlled by local chemistry. In aqueous phase the oxidants ozone, hydrogen peroxide and oxygen catalysed eventually by metal ions contribute to the oxidation. The dry deposition in the model is parameterized for different land types. Both in-cloud and sub-cloud scavenging are considered for wet deposition.

The simulations use the EMEP-MACC (Monitoring Atmospheric Composition and Climate) model configuration. The horizontal resolution of the model simulations is 0.25° (longitude) x 0.125° (latitude). There are 20 vertical layers up to about 100 hPa, with the lowest layer around 90 meters thick. The model is driven by meteorology from the European Centre of Medium-Range Weather Forecasts (ECMWF) in the MACC model domain (30° west to 45° east and 30° to 76° north). Iceland is in the upper ~~northwestern~~north-western corner of the domain, which implies losses of sulphur from the regional budget terms in sustained southerly and easterly flow regimes. The meteorology fields used have been accumulated in the course of running the MACC regional model ensemble forecast of chemical weather over Europe (<http://macc-raq-op.meteo.fr>), of which the EMEP/MSW model is part of. For our hindcast type simulations here, only the fields from the first day of each forecast are used. The meteorology is available with a three hourly interval. All model simulations are run from September through November 2014.

Emission from the Holuhraun fissure is set to a constant 750 kg/s SO<sub>2</sub> (65 kt/d) for the entire simulation. ~~from the total 2.0 ± 0.6 Tg SO<sub>2</sub> emitted in September estimated in Schmidt et al. (2015).~~ For all model runs the anthropogenic emissions are as standard for our EMEP MACC model configuration. Table 1 shows an overview of the four different model runs that are used in this study. ~~For the control run called ctr\_~~The column height observed both at ground and airborne instruments, varied during the eruption (Schmidt et al., 2015), the mean height was however around 3 km over the period. For the basic run called bas\_hol, volcanic emissions at Holuhraun are distributed equally from the ground up to a 3 km emission column height. To test the sensitivity towards emission height, two additional model simulations are done, ~~low\_hol and.~~ One simulation where the volcanic emission is distributed from the ground up to 1 km called low\_hol, and a simulation where the volcanic emission is distributed between 3km and 5 km called high\_hol. To derive the impact purely due to the emissions from Holuhraun, a simulation with no Holuhraun ~~fissure emissions is used, called no\_hol.~~ emissions is performed, called no\_hol. Sensitivity runs with an almost doubled constant emission rate of 1400 kg/s, and a time varying emission term given in Thordarson and Hartley (2015) were also studied. These resulted in an almost linear increase in concentrations and deposition, and did not compare better to observations and will therefore not be presented here. The sensitivity to height of the emission appeared to be more important and is shown here in more detail.

Anthropogenic SO<sub>2</sub> emissions in the model are described in Kuenen et al. (2014). There is a yearly total SO<sub>2</sub> emission of 13.2 Tg/a corresponding to 2009 conditions, the same year that is used in the reference MACC model configuration. The difference to actual 2014 conditions is assumed to be unimportant here. The inventory includes 2.34 Tg/a SO<sub>2</sub> in yearly ship emissions over the oceans. Over the continents the yearly emissions are 5.08 Tg/a SO<sub>2</sub> for the 28 EU countries, and 5.53 Tg/a SO<sub>2</sub> for the non-EU countries in the MACC domain (including Iceland) covered by the MACC domain.

## 2.2 Observations

The satellite data used in this study stem from the Ozone Monitoring Instrument (OMI) aboard NASA AURA (Levelt et al., 2006). The satellite was launched in July 2004 as part of the A-train earth observing satellite configuration and follows a sun-synchronous polar orbit. The OMI measures backscattered sunlight from the Earth atmosphere with a spectrometer covering UV and visible wavelength ranges. Measurements are therefore only available

during daytime. The background SO<sub>2</sub> concentrations are often too low to be observable, but increases in SO<sub>2</sub> from volcanic eruptions can produce well distinguishable absorption effects (Brenot et al. 2014). Pixel size varies between 13 km x 24 km at nadir and 13 km x 128 km at the edge of the swath. OMI satellite data are affected by “row anomalies” due to a blockage affecting the nadir viewing part of the sensor, which affects particular viewing angles and reduces the data coverage. The zoom-mode of OMI reduces the coverage on some days. The coverage is also reduced by missing daylight, e.g. winter observations from high latitudes are absent. Therefore data from only the two first months from September until the end of October are used in this study.

The retrievals are described in Theys et al. (2015). The sensitivity of backscatter radiation to SO<sub>2</sub> molecules varies with altitude (generally decreasing towards the ground level) and therefore the algorithms use an assumed height distribution for estimating the integrated SO<sub>2</sub> column density. Since often little information is available at the time of eruption and the retrievals produce results daily (even for days with no eruption) an assumed a priori profile is used for the vertical SO<sub>2</sub> distribution. The satellite retrievals used here assume an a priori profile with a plume thickness of 1 km that is centred at 7 km, similar to the method described in Yang et al. (2007). ~~This may be~~ As found in Schmidt et al. (2015), this is too high for the Bardarbunga eruption, ~~since our simulations indicate that. Therefore, the plume was often situated much lower in the troposphere. Retrieved retrieved~~ SO<sub>2</sub> column densities may ~~thus~~ be too low. To compare the vertical column density (VCD) from the model to the one from satellite retrievals, the averaging kernel from the satellite has to be used. Each element of an averaging kernel vector defines the relative weight of the true partial column value in a given layer to the retrieved vertical column (Rodgers, 2000). Cloud cover also changes the averaging kernel and a spatio-temporally changing kernel is part of the satellite data product (an averaging kernel is provided for each satellite pixel).

To apply the averaging kernel on model data, the satellite data are regridded to the model grid so that those data from satellite pixels nearest to any given model grid point are used for that grid point. A smaller area than the whole model domain was chosen to study and compare to the satellite data, 30° west to 15° east and 45° to 70° north (red boxes in Figure 1). The Aura satellite does five overpasses over the domain during daytime, swaths are partly overlapping in the northern regions. For the grid cells where the swaths overlap, the satellite observations are averaged to produce daily average fields. There are also regions that are not covered by

satellite observation that will not be taken into account in the model data ~~postprocessing~~. postprocessing. To make comparable daily averages of the model data, the closest hour in the hourly model output are matched to the satellite swath time and only grid points that are covered by satellite are used. The profiles for the averaging kernel in the satellite product are given on 60 levels, the values from these levels are interpolated to model vertical levels. The new adjusted model VCD is then calculated by multiplying the interpolated averaging kernel weights to the SO<sub>2</sub> concentration in each model layer, integrating all layers with the height of each model layer.

Because of noise in the satellite data small retrieved VCD values are highly uncertain. A threshold limit is sought to identify those regions that have a significant amount of SO<sub>2</sub>. Standard deviation for the satellite data is calculated over an apparently SO<sub>2</sub> free North Atlantic region (size 10 x 15 degrees lat lon respectively), and is found to be around 0.13 DU. Effects of varying cloud cover are ignored. An instrument detection limit is three times the standard deviation of a blank, so we assume that with a threshold value set to 0.4 DU we exclude satellite data below detection limit. Any grid point with a value over this threshold in the satellite data is used along with the corresponding model data. Daily mass burdens for the North Atlantic region are calculated by summing up all the SO<sub>2</sub> VCD in the grid cells above the threshold. One DU is  $2.69 \cdot 10^{20}$  molecules per square metre, which corresponds to a column loading of 28.62 milligrams SO<sub>2</sub> per square meter (mg/m<sup>2</sup>).

~~Station data~~ Data of SO<sub>2</sub> and PM<sub>2.5</sub> surface concentrations are collected by the European Environment Agency (EEA) through the European Environment Information and Observation Network (EIONET). We make use of two preliminary subsets of this data, one obtained from work within the MACC project to produce regular air quality forecasts and reanalysis, (only SO<sub>2</sub>), and a second one obtained from EEA as so called up-to-date (UTD) air quality data base, state spring 20152016. The two different subsets cover observation data from different countries, and have not yet been finally quality assured at the time of writing this paper. We use only station data, which contain hourly data, ~~however~~. However, there are missing data and some stations have instruments with high detection limits making it difficult to create a continuous measurement series with good statistics. Therefore, in this study some outstanding episodes with high concentrations of SO<sub>2</sub> are analysed. ~~Model data are picked consistently from gridded hourly data at model surface level~~ For the first six day period between 20th and 26th September, high concentrations of SO<sub>2</sub> were measured over Great Britain and countries

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further to the south. For the second six day period, a month later (20th to 26th October), the plume was also detected over Great Britain, but was transported further east towards Germany. For the last plume studied, lasting from 29th October to 4th November, the volcanic emission was transported eastward to the coast of Norway and countries to the south. Recent daily deposition data are taken from the EBAS data base (ebas.nilu.no) for those stations where the data are already available. Model data to represent the station values are picked from hourly data at model surface level in the grid cell where the station is located.

### 3 Results

#### 3.1 Comparison to satellite data

Observations by satellite provide information about SO<sub>2</sub> location and column density. Figure 1a shows as an example the VCD from the OMI satellite overpasses on 24 September. Fig. 1b and Fig. 1c show the modelled and the adjusted VCD from the ~~control~~basic run (~~bas\_hol\_ctrl~~). The observed satellite SO<sub>2</sub> cloud and the model simulated SO<sub>2</sub> cloud show similar shape and location. The adjusted model column densities are smaller than the original model VCDs. More weight is given by the averaging kernel to model layers higher up, close to the reference height of ~~7 km~~7 km, where there is less SO<sub>2</sub> in our case, with emissions and transport happening in the lower part of the troposphere. The reduced column densities are more comparable to the column densities observed by the satellite, ~~there~~. There are however some differences of where the maximum column densities are located.

A quantitative comparison is attempted here by integrating all satellite - and corresponding model data - above the North Atlantic, between Iceland and Europe, into daily mean column loads. Figure 2 shows time series from September to October of daily satellite coverage and daily mass burdens considered over the area where satellite VCD values exceed the 0.4 DU detection limit as explained above. The area covered by satellite observations at the beginning of the period is around 70 percent of the domain used here (red boxes in Fig. 1). Towards the end of the period, the satellite coverage is only around 40 percent because of the increasing solar zenith angle (a satellite zenith angle cutoff of 75° is used for the satellite data). On some days, the satellite cover is even lower because of the OMI zoom mode. The percentage of the satellite data that is above the detection limit is low over the entire two month period, only reaching around ten percent at the end of September and at the beginning of October.

On most days, the satellite daily mass burdens are above the model value, not including the days where the zoom mode minimizes the coverage. The average mass burden adjusted to the 7 km reference height for satellite data are 11.17 kt SO<sub>2</sub> for satellite and 8.72 kt SO<sub>2</sub> for the model. The highest values are at the beginning of the period, 42.11 kt SO<sub>2</sub> for the model data on 7 September, and 37.42 kt SO<sub>2</sub> 20 September for the satellite data. The daily values are decreasing over time, ~~for both observed and model mass burdens. Especially~~ during October ~~the values are declining~~. At the same time the satellite coverage is decreasing. To further investigate whether the increasing solar zenith angle is responsible for the increasing bias of the simulated versus observed VCDs, a new domain further south is used. All ~~that~~the area where satellite observations may be possible until the end of October (61.25° north) is used to calculate another set of daily column loads for satellite and model data (see Fig. 2c). Satellite coverage in this southerly domain is not decreasing over time, but it is also not covering Iceland, so the SO<sub>2</sub> from Holuhraun needs to be transported south to be detected. The plume is transported south four times over the two-month period as the peaks in column load values show. In this southerly area the daily accumulated mass burdens are similar in September and in October, supporting the idea that the decrease in mass burden in Fig. 2b is due to reduced satellite coverage. Taking into the account the area in which the satellite observed SO<sub>2</sub> above detection limit, the satellite average column loads are calculated as to around 70 mg/m<sup>2</sup> for the start of the period and on 19 September, model values are lower. Also the peaks in the middle of October in Figure 2b have a satellite average column value at 62 mg/m<sup>2</sup>.

Percentile values from the distribution of the daily mass burden in September and October 2014 from all the three model simulations, original and kernel weighted are shown in Fig. 3. The kernel weighted model data can be directly compared to the percentile characterisation of the satellite data. As illustrated in Fig. 1, there is a clear decrease in the column load values before and after the averaging kernel is applied, because the SO<sub>2</sub> plume was found much below 7 km altitude. The differences between the three model simulations however change before and after the satellite kernel is applied. For the original model data, the model simulation with emissions in the lowest kilometre (low\_hol) has the highest daily mass burden values, while the run with the emission highest in the atmosphere (high\_hol) exhibits a lower mass burden than the two other. The higher values in the low\_hol simulation can be explained by less wind and dispersion at low altitudes and thus a more concentrated SO<sub>2</sub> cloud than in the two other model simulations. After the averaging kernel is applied to the



model data, the high\_hol model simulation has the highest daily values compared to the other two model simulations. High values in satellite data, and model data with kernel profiles applied reflect high concentrations and/or volcanic SO<sub>2</sub> at high altitudes.

Comparing the satellite data to the kernel weighted model data; the satellite 75<sup>th</sup> percentile is higher than the model 75<sup>th</sup> percentile. The median for the et#bas\_hol, low\_hol and high\_hol daily mass burden are 7.38 kt, 4.43 kt and 8.34 kt respectively, for satellite the mass burden median value is 7.03 kt. The satellite data therefore have higher maximum values that results in the higher average values and the 75<sup>th</sup> percentile, most of the satellite daily mass burden values are however around the model data for the et#bas\_run. From all the model simulations, with different emission heights, the et#bas\_run is the most similar to the satellite data over the first two months.

### 3.2 Surface concentrations

SO<sub>2</sub> from the volcanic eruption on Holuhraun was measured at several surface stations during the period. Three different episodes with clear peaks in observed concentrations at stations around Europe are described in the following paragraphs. Exemplary comparisons are shown and additional comparisons at other stations are available in the supplementary material.

Figure 4 shows hourly time series for two stations over Great Britain and France from 20 September to 26 September. ~~On 21 September 16 UTC, high~~Concentrations of 44.25 µg/m<sup>3</sup>, SO<sub>2</sub> concentrations were measured 21 September 16 UTC at ~~the station in Great Britain. The~~a station is situated in Manchester (53.48°N and 2.24°W) near the west coast of Britain. None of the three model simulations exhibits exactly the same values as observed. Although the model simulations do not reach the observed maximum values, the model field shows areas south of the station nearby Manchester, where the SO<sub>2</sub> concentrations only due to the volcanic eruption are around 50 µg/m<sup>3</sup>. Interestingly, the agreement of the model derived volcanic SO<sub>2</sub> time series is better in agreement with measurements than the total simulated SO<sub>2</sub> concentration (grey curve), indicating that the model may not resolve SO<sub>2</sub> transport from nearby pollution sources and that the station for these days is rather representative of long range transported SO<sub>2</sub>. Observed PM<sub>2.5</sub> concentration at the station shows, that over the period, the highest concentration (52.1 µg/m<sup>3</sup>) – probably anthropogenic - is measured at the start of the period, before any volcanic sulphur contribution is simulated by the model. The next day, the plume has moved further south over France. ~~The French, the~~ station is situated

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on the west coast of France in Saint-Nazaire- (47.25°N and 2.22°W). The measurements show three peaks over three days, with the highest one (38  $\mu\text{g}/\text{m}^3$ ) measured 12 UTC 23 September. All the three model simulations have the peak concentrations earlier than the observed, and the concentrations from the model are lower than observed. The three simulations do however show increased concentrations at the site due to the volcanic eruption over the three days. The map shows that large parts of France had an increase in  $\text{SO}_2$  surface concentrations during this time.

Figure 5 shows the time series for three stations over Scotland and Germany a month later, from 20 to 26 October. The high\_hol simulation shows low concentrations over the Scottish Grangemouth station- (56.01°N and 3.70°W), but the ~~ctrl~~bas\_hol and low\_hol have a plume with high concentrations over the station on 20 October. There are no measurements at this time to compare the model values to. The timing of the second plume 21 October for the two models is a few hours early and the modelled concentrations higher than the 6.09  $\mu\text{g}/\text{m}^3$  observed, especially for the low\_hol simulation. The map shows a narrow plume from Iceland south to Scotland and the station lies on the edge of this plume. On 22 October, the volcanic  $\text{SO}_2$  is measured at stations in Germany. Figure 5d shows the plume reaching from Iceland into the North Sea, transported east and south compared to the situation from the day before. The two stations Kellerwald (51.15°N and 9.03°E) and Bremerhaven (53.56°N and 8.57°E) experience the plume differently. While for Bremerhaven the peak observed (41.0  $\mu\text{g}/\text{m}^3$ ) is short in duration, the peak lasts for one day at Kellerwald- with an observed maximum of 10.2  $\mu\text{g}/\text{m}^3$ . The map ~~shows~~shows that the plume is narrow for all three stations; and the gradient between where there is no Holuhraun contribution and the maximum concentration is strong. ~~At Kellerwald station, the low\_hol simulation has the highest concentrations at the beginning of the plume and the high\_hol simulation is highest at the end of the plume. The ctrl\_hol simulation has the most comparable concentrations to the observed ones, although all the models runs have values that are too high. For the Bremerhaven station, the observed peak is earlier than the modelled, but all the model runs match the last hours of the plume.~~

A third plume is illustrated in Fig. 6 over Northern Europe, occurring from the end of October to the beginning of November. Figure 6a shows the measured  $\text{SO}_2$  concentrations at a station in Oslo, Norway- (59.92°N and 10.76°E). There are four peaks measured from 29 October to 31, the highest one on 29 October- (50.4  $\mu\text{g}/\text{m}^3$ ). The models runs show contribution from Holuhraun  $\text{SO}_2$  over the same three days, but do not reach the high measured concentrations,

especially the first plume is underestimated. On October 30, the plume is transported south east to Poland. The Polish station in Sopot (54.43°N and 18.58°E) experiences a short peak that the model simulates to happen a few hours earlier. ~~The etl, With the bas~~ hol simulation has the most comparable concentrations.

Figure 7 shows wet deposition for the whole three month period at the Kårvatn station (62.78°N and 8.88°E) and the west coast of Norway. There are high levels, both observed and modelled during the last part of September. The model exhibit high values on 27 September, while the observed deposition is spread out over several days. Summed over the whole period, the observation has 15.9 gS/m<sup>2</sup>y while the bas model simulated 19.98 gS/m<sup>2</sup>y. Comparisons at other stations in Norway also show the same results (appendix).

Transport to Europe is caused by northerly and north-westerly winds. For the first plume, where the model shows low concentrations compared to the observations, there had been southerly winds a time before strong northerly winds transported the SO<sub>2</sub> cloud south over Great Britain and France. Compared to the other two episodes, the SO<sub>2</sub> surface concentration due to Holuhraun are higher over a larger area during this episode. The difficulty of the model to simulate the SO<sub>2</sub> transport correctly depends on the uncertainty in the emission term, the meteorology fields, the chemical reactions and deposition. Overall the comparison ~~at the stations and with the~~ to observations shows, taking into account satellite and station data ~~indicates~~, that the ~~etl~~bas\_hol model simulation, ~~matches best with the observed satellite column burdens, their time evolution and for some stations~~ with the ~~assumption that emissions occurred between 0 to 3 km, performs best~~ magnitude and timing of the observed peaks.

### 3.3 Effects of the eruption on European pollution

The results above show that, although the Holuhraun eruption released large amounts of SO<sub>2</sub>, the stations in Europe often measured the increase in SO<sub>2</sub> ~~concentrations~~concentration as short peaks (Grislason et al. 2015, Schmidt et al. 2015). The model makes it possible to find a more general view of the impact in the European air quality due to the volcanic emissions. Table 2 summarizes the model results for Europe. ~~Only grid~~Grid cells ~~covering one of~~covered by the ~~34~~ countries mentioned are ~~considered when used for~~ calculating the results shown in the table, ~~the~~. The emission (from anthropogenic sources), concentration and deposition over the oceans are not ~~studied~~included. Since a large part of the deposition and concentration

1 increase occurs downwind and to isolate the effect on Iceland itself close to the emission  
2 point, the deposition and concentrations over Iceland ~~is~~are given in brackets.

3 The Holuhraun emission estimate used in this study releases over 4.5 times the anthropogenic  
4 emission from the 31 countries (not including ship emissions). The anthropogenic emissions  
5 from Iceland are only 18 kilotons, the SO<sub>2</sub> emissions from Iceland increase by ~~over~~more than  
6 300 times.

7 Over the three months, there is 1.32 times more SO<sub>x</sub> wet deposition for the ~~control~~basic run  
8 with Holuhraun emission than the MACC reference with no Holuhraun emission. ~~The wet~~Wet  
9 deposition over ~~Iceland and the rest of~~ Europe is dependent on the emission height. The  
10 simulation with the emission highest in the atmosphere (high\_hol) has the highest  
11 contribution to the rest of Europe, ~~while less than half of the wet deposition falls on Iceland~~  
12 ~~compared to the other two runs~~. For dry deposition, the ten percent increase over Europe is  
13 about the same for all the three model simulations with Holuhraun emissions. ~~For~~Close to the  
14 source, over Iceland ~~however, the SO<sub>x</sub> dry deposition is~~levels are very dependent on  
15 ~~emission~~the emissions height, especially for dry deposition.

16 Figure ~~7~~8 shows the total deposition over Europe for the standard MACC model simulation  
17 with no Holuhraun emission (no\_hol), the ~~control~~basic model simulation (~~ctrl~~bas\_hol), and  
18 the percent increase ~~for~~between these two model runs. ~~For the no\_hol simulation, the highest~~  
19 ~~depositions are over central and Eastern Europe, while the areas with the lowest deposition~~  
20 ~~are over Iceland, northern Scandinavia and over the Alps. These are also the areas~~Areas that  
21 experience the highest percent increase ~~in addition to the northern part of Scotland~~are also  
22 areas that have low levels in the model simulation with no emission at Holuhraun. Due to the  
23 Holuhraun emissions Iceland has the highest SO<sub>x</sub> deposition in Europe, and the coast of  
24 northern Norway shows depositions on the same level as ~~Eastern Europe. the more polluted~~  
25 eastern Europe. Even though the previous section indicated that the model has higher wet  
26 deposition levels in northern Norway than observed, it also showed that it is very likely that  
27 the observed increases in SO<sub>x</sub> deposition levels are due to the Holuhraun emissions.

28 The averaged SO<sub>2</sub> surface concentration over Europe is under normal ~~condition~~conditions  
29 higher than over Iceland. ~~For, the simulations with Holuhraun volcanic~~ emission caused the  
30 concentration level over Iceland to increase ~~over~~by a factor of 177 (for the low hol  
31 simulation). Over the rest of Europe the increase is around the same for all three Holuhraun  
32 simulations, even though the time series showed that the different simulations. ~~The ctrl\_hol~~

1 ~~and ctrl\_low simulation give high increases over Iceland, while for the high\_hol simulation,~~  
2 ~~the had peaks arriving at often different times. On average concentration over Iceland is close~~  
3 ~~to the rest of, however, vertical mixing has levelled off initial differences in emission height~~  
4 ~~when volcanic plumes arrive in~~ Europe.

5 The increases in PM<sub>2.5</sub> concentrations are due to increased sulphate production from volcanic  
6 SO<sub>2</sub>. ~~Dry production is due to SO<sub>2</sub> reacting to OH, while wet production occurs in cloud~~  
7 ~~droplets. PM<sub>2.5</sub> concentrations are a PM<sub>2.5</sub> is a~~ collection of all ~~aerosol~~ aerosols under 2.5 µm,  
8 ~~and sulphur is only a part of the therefore the increased sulphate is changing total~~ aerosol  
9 mass. ~~For PM<sub>2.5</sub> concentrations, the relatively little. The~~ table shows that Iceland has a lower  
10 average concentration than the rest of Europe for all the four runs, even though Iceland is the  
11 ~~contributor~~ origin to the increase in aerosol pollution levels. The high\_hol model simulation  
12 has a higher increase in PM<sub>2.5</sub> concentration over Europe than the two other simulations.  
13 ~~Especially the The low\_hol simulations have high~~ simulation finds highest sulphate and SO<sub>x</sub>  
14 deposition on Iceland itself, and possibly over the nearby ocean, that will lead to a lower  
15 contribution to ~~the PM<sub>2.5</sub> increase~~ pollution levels over the rest of Europe.

16 The distribution of PM<sub>2.5</sub> from the no\_hol and ~~ctrl~~bas\_hol simulation, plotted in Figure 89,  
17 shows the same polluted and clean areas as in Fig. 7. ~~The percent-8, although the~~ increase is  
18 ~~not as high as for the deposition, but the areas are similar. There is a high increase over~~ lower.  
19 Over north-west Norway and northern Norway, ~~where~~ the increase is over 100 percent.  
20 Figure 8b ~~still~~ 9b shows that although the percentage increase is high, the PM<sub>2.5</sub> concentrations  
21 in these areas are still among the least polluted in Europe. The high deposition levels in this  
22 region indicate that some of the PM<sub>2.5</sub> is scavenged out.

23 WHO recommends a 24 hourly average mean concentration level of 25 µg/m<sup>3</sup> for PM<sub>2.5</sub> not to  
24 be exceeded over three days over a year (WHO, 2005). Figure 9a 10a shows that over the  
25 Benelux region, ~~Northern~~northern Germany and ~~Northern~~northern Italy this limit value is  
26 exceeded by up to ten days during the three months studied. As the previous plot showed,  
27 these are regions with high average PM<sub>2.5</sub> concentrations. Because the daily concentrations  
28 are already high, any increase in days in the model ~~ctrl~~bas\_hol simulation due to the  
29 Holuhraun emissions is also occurring in these regions, and the areas with the highest percent  
30 increase does not experience any days over the limit. The Figure also shows that Northern  
31 Ireland experienced up to two exceedance days due to the volcanic eruption.

## 4 Discussion

The variances between the satellite model data and the satellite observations can be due to several factors. a) The model emissions flux may be under or overestimated compared to the real emissions, model VCDs are therefore too low / too large compared to the observed ones. b) The areas within which the column mass are constructed depend on the threshold VCD value and the satellite data, so the values in the model depend on the position of the observed SO<sub>2</sub> cloud. If the simulated plume is displaced into an area where the satellite does not show any useful signal, then this part of the model plume is ignored and may lead to underestimates of the model. c) The presence of clouds can increase the uncertainty of the satellite retrieval.

~~d) The unknown real height of the SO<sub>2</sub> plumes~~ Schmidt et al. (2015) presents IASI (Infrared Atmospheric Sounding Interferometer) plume heights between 5.5 km to 1.6 km derived from an area of 500 km around the volcanic location, and a mean IASI centre of mass height between 2.7 km to 0.6 km. The fluctuating real height of the SO<sub>2</sub> plume may introduce additional bias between model and satellite VCDs.

Schmidt et al. (2015) presents a comparison between model, satellite and ground observations for September. Mass burdens from OMI are derived using observed plume heights from the IASI instrument on the MetOp satellite. The model NAME (Numerical Atmospheric-dispersion Modelling Environment), a Lagrangian model, is run for September with sensitivity runs testing both emission height and emission flux. Comparing with the two satellite data sets, the model simulation with a plume height of 3 km and doubled emission flux (~1400 kg/s) matches well with the OMI satellite data for the first days, while for the rest of September the model simulation with emission similar to the constant emission used here matches better (~700 kg/s). In this study, since the model data is weighted with the averaging kernel before compared to the satellite data the values are lower, because the assumed plume height is 7 km. Both methods show however that for the first days, the satellite had higher values than the model for the first days and at the end of September. Model simulations with higher emissions showed better comparison during the first days of September, but overall the height of the plume is more important for the satellite comparison.

Our Holuhraun emission term in the three model simulations is constant throughout the simulations both with respect to emission height and emission flux. Maximum fluxes of 1300 kg/s were reported by Barsotti (2014), and Gislason et al. (2015) estimated a 2.5 times the average emission term during the first two and a half weeks of the eruption. The assumption

1 of a constant emission term is thus certainly a simplification. The emission height is also  
2 variable, dependent on initial volcanic eruption characteristics and meteorological conditions  
3 like wind speed and stratification (Oberhuber et al. 1998). A better source estimate for the  
4 eruption is beyond the scope of this study; however the fluctuations in flux magnitude and  
5 emission height can explain some of the differences between observed and simulated  
6 concentrations, especially at the beginning first days of September in the satellite comparison.

~~Ialango et al. (2015) found that the SO<sub>2</sub> plume from Holuhraun was detectable with a Brewer  
instrument in Finland and compared the measurements to satellite observations from OMI  
(Ozone Monitoring Instrument) and OMPS (Ozone Mapping Profiler Suite). From comparing  
the ground measured SO<sub>2</sub> to the satellite data, the satellite products with an a priori profile  
placing the SO<sub>2</sub> in the planetary boundary layer gave the best agreement. The reduction in  
column loads from applying the averaging kernel seen in this study leading to reasonable  
agreement with the satellite VCDs also shows that the SO<sub>2</sub> was situated well below the 7 km  
altitude. Further comparison of the modelled SO<sub>2</sub> vertical distribution to measured one, e.g.  
from IASI, is needed to understand the impact of any bias in modelled vertical distribution on  
the comparison to satellite derived VCDs. Our sensitivity runs indicate considerable  
sensitivity of the estimated amount of SO<sub>2</sub> in the North Atlantic area to the vertical  
distribution of the SO<sub>2</sub>. This essentially prevents us from using the satellite data to make a  
more quantitative inverse judgement on the emission strength.~~

~~Schmidt et al. (2015) presents a comparison between model, satellite and ground observations  
for September. Mass burdens from OMI are derived using observed plume heights from the  
IASI (Infrared Atmospheric Sounding Interferometer) instrument on the MetOp satellite. The  
model NAME (Numerical Atmospheric dispersion Modelling Environment), a Lagrangian  
model, is run for September with sensitivity runs testing both emission height and emission  
flux. Comparing with the two satellite data sets, the model simulation with doubled emission  
flux (~1400 kg/s) matches well with the OMI satellite data for the first days, while for the rest  
of September the model simulation with emission consistent with this paper matches better  
(~700 kg/s). The satellite comparison presented here shows that although the satellite data  
have higher daily mass burden values for most of the first days, it is not evident that the  
emission term on average is too small. The observed plume height presented in Schmidt et al.  
(2015) by IASI measurements also supports our ctrl\_run emission height distribution between  
the ground and 3 km.~~

Surface concentration comparisons presented in this study and in the supplementary material show that the volcanic SO<sub>2</sub> was observed as short singular peaks lasting from a few hours to several peaks over a short set of days. The biggest difference for the three studied plumes is for the first one during 20 to 26 September over UK for the Manchester (GB0613A) in Great Britain and Western Europe the Saint-Naizaire station (FR23181) in France, with up to a factor of four differences between simulated and measured concentrations ~~at several of the stations. But both.~~ Both the measured and simulated concentrations during the September event were higher than the two later events, pointing to a different transport of SO<sub>2</sub> in the first event, and not only higher emissions. ~~Schmidt et al. (2015) also presented a model comparison of observed and model concentration for these days, and the results show the same as seen in this study. Even for the NAME model simulation with double emission show smaller concentrations at the stations presented in Schmidt et al. (Higher emission fluxes are also not supported by the satellite comparison over these days either. Changes in emission flux for the EMEP/MSC-W have been shown to have an almost linear change in concentrations (not shown here); even with doubled emissions during this event the model would still simulate concentrations and burdens well below those observed. Station data presented in Schmidt et al. (2015) for these days show the same results, indicating that the models and meteorology had difficulties representing this period.~~

The discrepancies between the model and observations, especially for the station data show that the values presented in Table 2 contain error. Especially the model surface concentrations are low compared to observations; however the map plots show, that sometimes modelled concentrations nearby the stations reached observed levels. The area averaged concentrations presented in table 2 may therefore be close to the real concentration increase. A more thorough study of longer time series with deposition and concentration trends is needed to estimate better the increase in SO<sub>2</sub> concentrations due to the eruption at the stations.2015).

The results in this study show that the sulphur depositions from September to November over Northern Norway were at the same levels as the most polluted regions in Europe. Emission ceilings aim set by the Gothenburg Protocol was to reduce the SO<sub>x</sub> emissions by 63 % by 2010 compared to the 1990 levels (EMEP, 2015). Most countries have accomplished these reductions, and the sulphur deposition levels over Europe have decreased. The Holuhraun eruption changed the picture in some areas. Comparing observed deposition levels at Tustervatn station in central Norway, the simulated deposition is higher than the yearly

Formater



observed averages since 1980. Monthly observed values at this station during the 2011 Grimsvötn eruption show almost as high values as the ~~ctrlbas~~ hol simulation. The ~~increase in SO<sub>x</sub> deposition at Birkenestime series from the Kårvatn station in Southern Norway is negligible~~ also shows that the increases are due to the Holuhraun volcanic eruption. Northern Norway is more susceptible for volcanic impact because of the geographical position, in addition to high frequency of precipitation on the western coast of Norway. Comparing the mean deposition levels over the three months in 2014 over Norway to model simulations with emissions from previous years, they are double to the early 1990s (EMEP, 2015). Southern Norway experienced a sulphur deposition decrease of 40 % from 1980 to 1995 due to emission abatement in Europe (Berge et al. 1999). The highest contributors to high deposition levels over Southern Norway were the UK and Germany (18 % and 15 % respectively). Norway also experienced in 2014 a high percent increase in PM<sub>2.5</sub> concentrations. The PM<sub>2.5</sub> levels over Scandinavia are low, and a small increase in the concentrations leads to high percent increases. The increase over land shows a similar pattern as the results found in Schmidt et al. (2011) for a hypothetical Laki eruption. Even though the highest increase is over Scandinavia and Scotland, the concentrations are too low to exceed the 25 µg/m<sup>2</sup> limit. Already polluted regions like the Benelux region experience more days with exceedances as well as North Ireland.

## 5 Conclusions

The increase in ~~emitted~~ SO<sub>2</sub> ~~to the atmosphere~~ caused by the volcanic eruption at Holuhraun were observed by satellite and detected at several stations over Europe- (Schmidt et al. 2015, Gislason et al., 2015). Model simulations with the EMEP/MSC-W model with emissions from Holuhraun over the period from September to November have been done to investigate the model capability to simulate such events, and also to study the impact of the increased emissions on concentrations and depositions over Europe.

The first two months of the model simulations are compared to satellite retrievals from OMI. The retrievals use an assumed plume height of 7 km. Averaging kernels from the satellite data are applied on the model data to compare the model data to the satellite. Because of the weighting, the satellite retrieved mass burden values are dependent on both vertical placement and amount of SO<sub>2</sub>. Two sensitivity model simulations with different Holuhraun emission height are compared to the satellite data together with the ~~control~~basic simulation. ~~The results show-After the importance of weighting the model data with the averaging kernel when~~



~~comparing the model is applied, the results are more comparable to the satellite VCD. The combined uncertainty in emission strength and height impact when comparing the satellite data to the model simulations makes data. The results also show that it is difficult to conclude which emission height is most realistic. if the discrepancies are due to the concentrations or the vertical placement.~~

The model simulations are compared to observed concentrations at stations over Europe for three different events with high concentrations measured at the stations due to the Holuhraun emissions. For all the events, the timing of the model peaks is well compared to the observed peaks in concentration. ~~For~~ There is a better timing in the two model simulations ~~with where~~ the emissions distributed are injected lowest ~~in into~~ the atmosphere, ~~a better timing can be seen~~ than for the sensitivity run with the highest emission height. Due to the transport of SO<sub>2</sub> during the first event, ~~both the model data and measurements~~ observed concentrations are higher than during the ~~two latter later~~ events. ~~The biggest, and the difference in concentration between observed models and simulated values observations is also seen largest. PM<sub>2.5</sub> concentration during this first plume reaching Europe event is comparable to observations.~~ Uncertainties in the model simulations increase by the length of transport, and some near misses of the narrow plumes can clearly explain differences between model and observation. Also, to make a better estimate of the model performance during the whole volcanic eruption, better quality checked station data is needed. Comparison between the model and wet deposition observations over Norway show significant and high contributions from the eruption, although the model over-predicts values at the station studied and other stations showed in the appendix.

~~The change~~ Studying the changes in pollution levels over Europe ~~due to the increase in emissions due to the volcanic fissure is studied. Of the parameters studied, SO<sub>x</sub> wet deposition showed the highest increase. in the model.~~ For the ~~control~~ basic simulation there is 32 % ~~times~~ more sulphate wet deposition than the model simulation with no Holuhraun emission over the 28 European Union countries, Norway and Switzerland. The regions that have the highest increase, apart from Iceland, are Northern Scandinavia and Scotland, regions that are among the least polluted in Europe. Especially the coast of Northern Norway, with a percent increase in total deposition of over 1000%, has shows levels ~~in 2014~~ equal to the most polluted regions in Europe. Compared ~~to measurements, the with observed levels are higher than the yearly averaged measured ones at since 1980 at the~~ Tustervatn ~~(Central station in~~

~~central Norway) since 1980 the 2014 model values are earlier only reached in the observations during the Grimsvötn eruption in 2011. Higher values measured at the Kårvatn station in 2014 on the coast of western Norway are due to the Holuhraun emissions.~~

Compared to model simulations with meteorology and emission from previous years, the mean deposition levels over Norway are double that of 1990.

The difference in SO<sub>2</sub> concentrations over Europe between the no\_hol and model simulations with Holuhraun emission are around 13 percent over the same 30 countries and increases occurs as short peaks in concentration levels from a few hours to some days. ~~Due to the underestimation seen at stations during September, the uncertainty of this number is large and the increase is possibly too small.~~ For PM<sub>2.5</sub> concentration, the increase is six percent~~-, and the model shows better agreement with station observations.~~ The biggest difference in percent increase ~~are~~~~is~~ seen over Scandinavia and Scotland, however these regions are among the cleanest in Europe, also with the added sulphur caused by the Holuhraun emissions. A lot of the sulphur is also deposited out over these regions by frequent precipitation. The areas that show increase in days with over 25 µg/m<sup>2</sup> PM<sub>2.5</sub> concentrations are already polluted. Even ~~though~~ with high ~~emission~~~~emissions~~ from the volcanic fissure at Holuhraun, the ~~increases~~~~increase~~ in pollution levels ~~are low~~ over Europe. is relatively small, with only transient episodes associated with high increases in SO<sub>2</sub> concentration.

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Formater

1 Table 1. Overview of model runs and the Holuhraun emission height assumptions and flux.

Model run name	Holuhraun layer into which SO <sub>2</sub> was injected in the model simulation	Holuhraun flux
<del>et#bas</del> _hol	0 - 3 km	750 kg/s
low_hol	0 - 1 km	750 kg/s
high_hol	3 - 5 km	750 kg/s
no_hol		0

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1 Table 2. Emissions, depositions and concentrations for the 28 European Union member states,  
2 Norway and Switzerland for the three months (September, October, November). Emissions  
3 and depositions are total over the three month period, concentrations are the mean over the  
4 period for the 31 countries. Numbers in brackets are the contribution from Iceland, for  
5 emission and deposition, the number represents the sum over Iceland. For concentration, the  
6 number represents the average over Iceland.

	no_hol	<del>et</del> bas_hol	low_hol	high_hol	<del>et</del> bas_hol/no_hol
Emissions SO <sub>2</sub> <del>(</del>	1 257	1 257	1 257	1 257	1
<del>]</del> [kilotons]	(18)	(5 980)	(5 980)	(5 980)	(5.68)
SO <sub>x</sub> Wet deposition	1 043	1 382	1 285	1 465	1.32
<del>(</del> [kilotons]	(11)	(1 122)	(1 491)	(472)	(2.37)
SO <sub>x</sub> Dry deposition	481	529	524	526	1.10
<del>(</del> [kilotons]	(4)	(151)	(409)	(8)	(1.40)
<u>Mean</u> SO <sub>2</sub>	1.39	1.58	1.56	1.56	1.13
<del>Surface</del> <u>surface</u> conc.	(0.59)	(38.95)	(105.91)	(1.81)	(66.17)
<del>(mean- [μg/m<sup>3</sup>])</del>					
<u>Mean</u> PM <sub>2.5</sub>	5.86	6.20	6.09	6.28	1.06
<del>Surface</del> <u>surface</u> conc.	(0.82)	(2.50)	(3.13)	(1.12)	(3.06)
<del>(mean- [μg/m<sup>3</sup>])</del>					

7



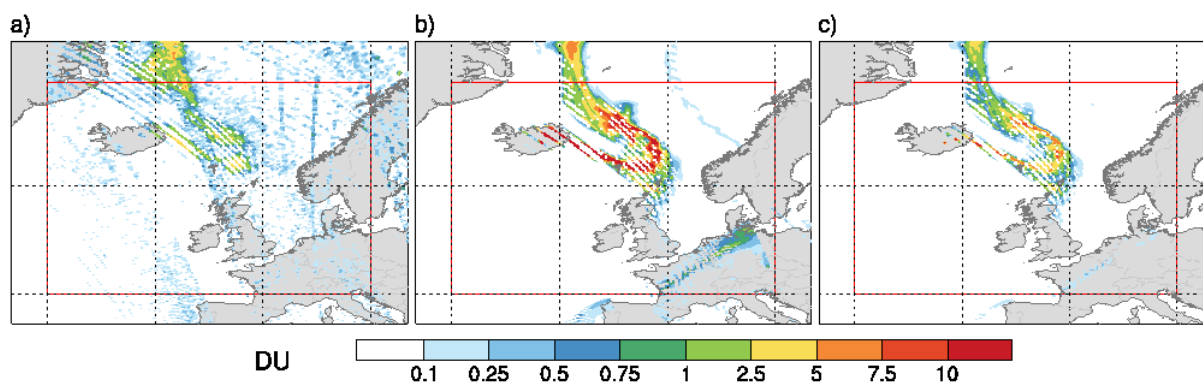
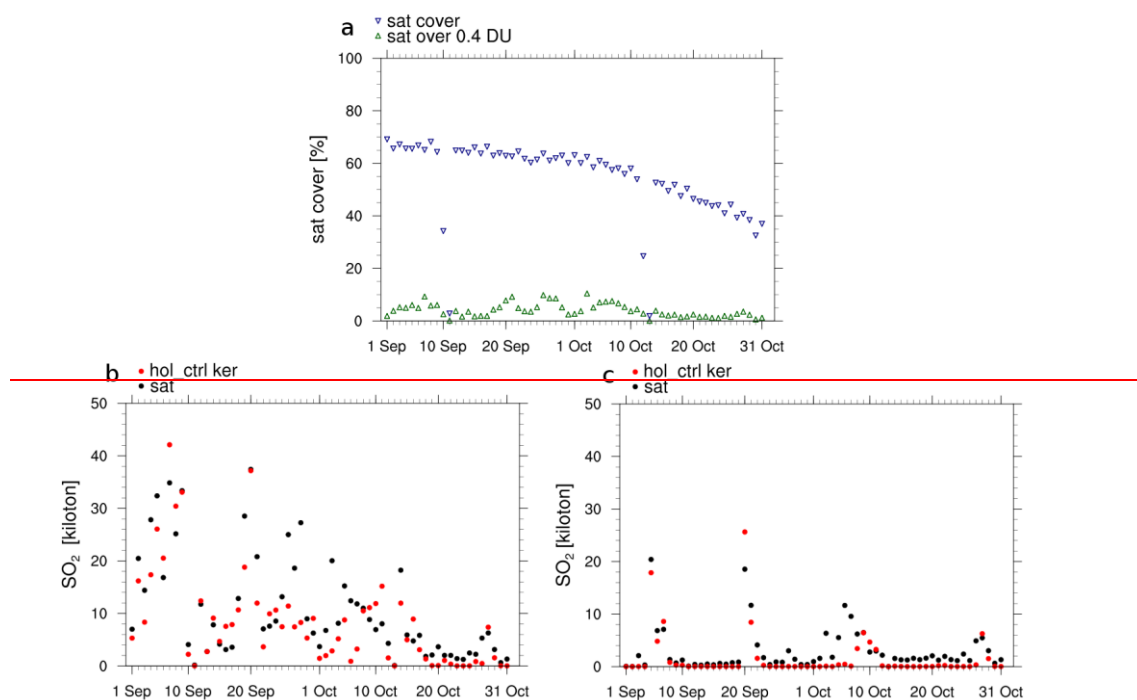


Figure 1.  $\text{SO}_2$  column density for a) the satellite swaths on 24 September, b) corresponding model data for the basic simulation from 24 September, and c) model bas\_hol data with averaging kernel applied from satellite data. The red box indicates the area where the satellite statistics in fig.2 are done.



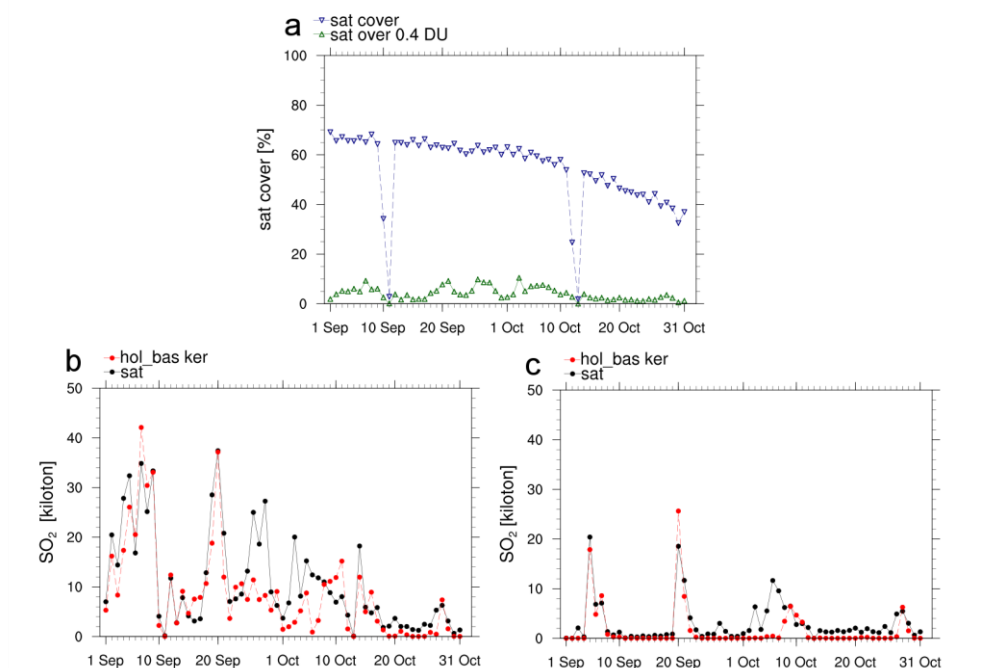


Figure 2. a) Daily time series of satellite observed area coverage (blue triangles) in percent of the total area of the domain used for the statistics (30 W - 15 E and 45 - 70 N, see fig 1). Green triangles show the percent of the area where satellite derived SO<sub>2</sub> is above 0.4 DU. b) Daily time series of mass burdens from satellite data (black dots) and from model control run (red dots) with averaging kernel applied, accumulated in consistent area. c) Shows the same as b) but over a smaller area just south of 61.15 degrees north.

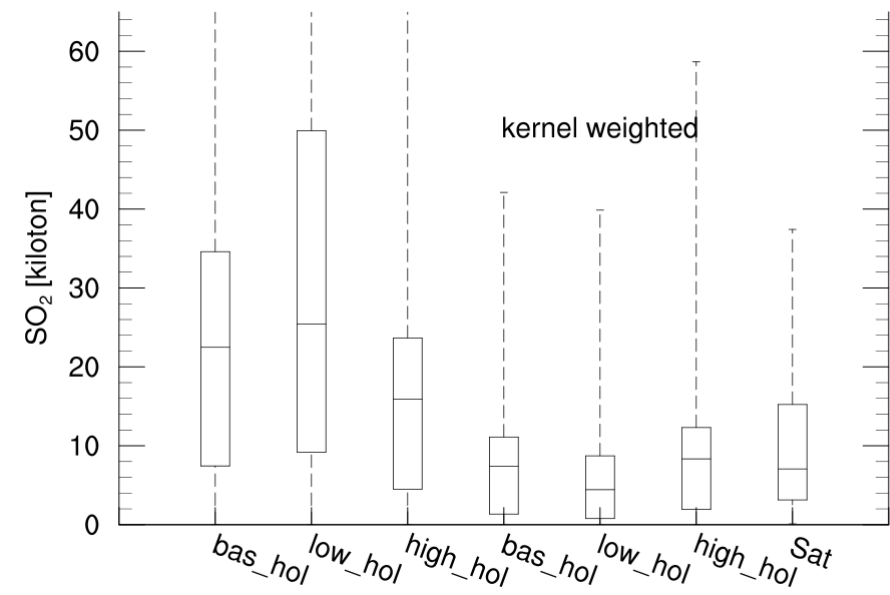
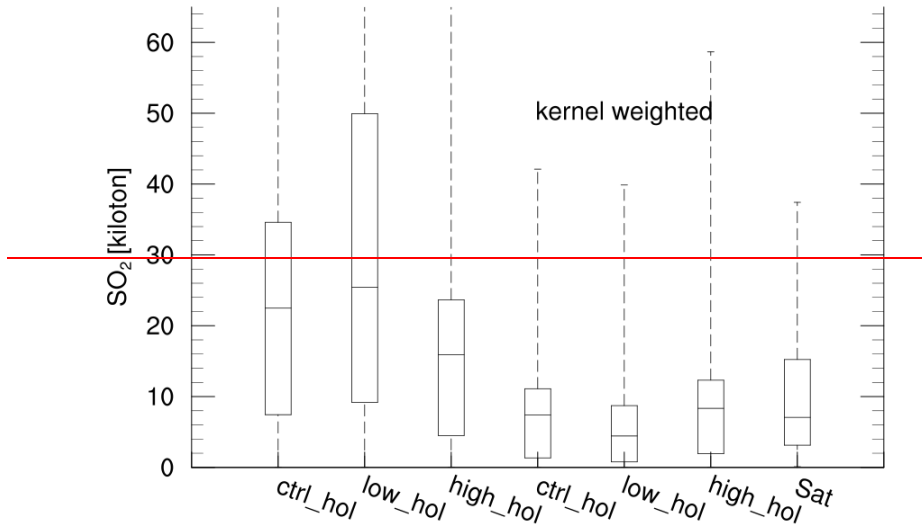
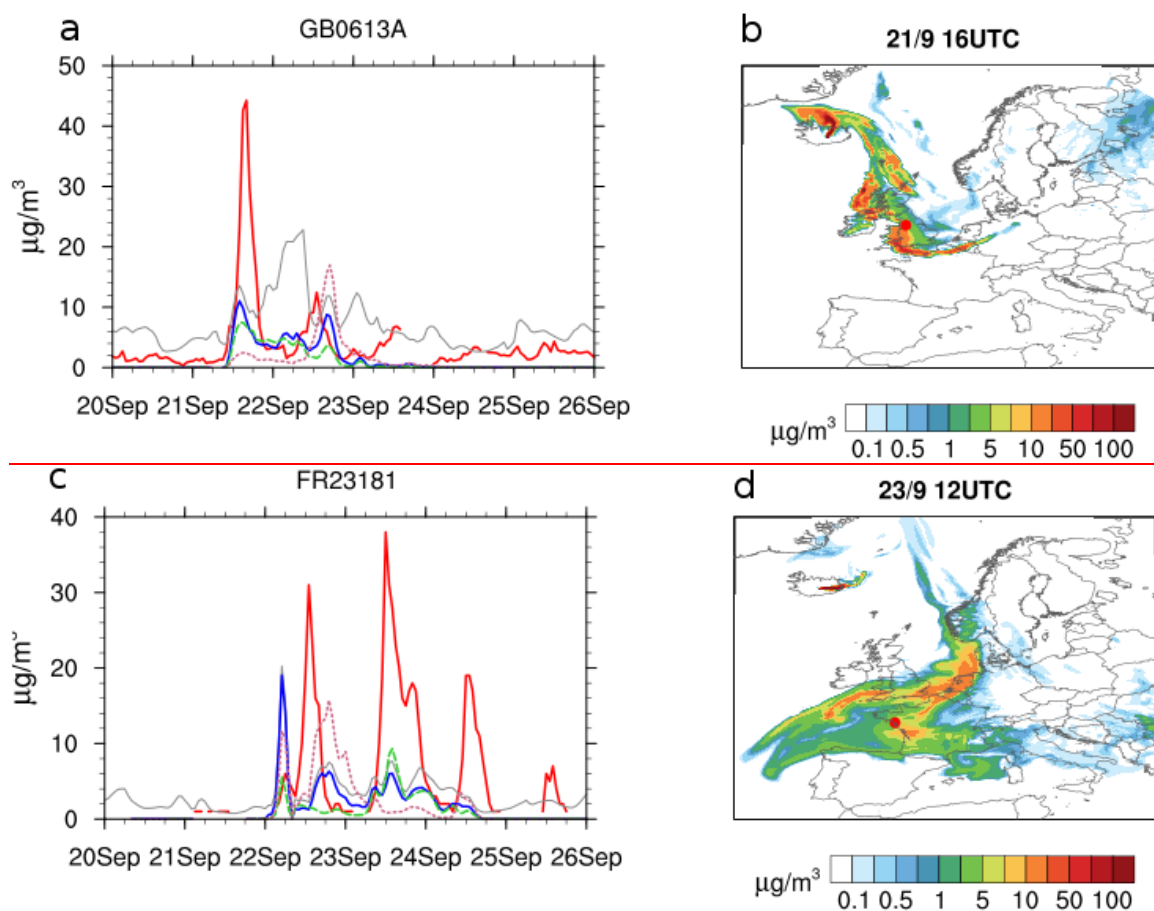


Figure 3. Distribution of mass burden derived from the 61 daily values (see fig 2) for the three model simulations, one for each of the three kernel weighted and the satellite data, in the area where satellite derived SO<sub>2</sub> exceeds 0.4 DU. The boxes shown represent the 25<sup>th</sup> percentile, the median, and the 75<sup>th</sup> percentile values, lower whiskers the minimum value and upper whiskers the maximum value.

~~Percentile statistics derived from the 61 daily mass burden values (see fig 2) for the three model simulations, each of the three kernel weighted and the satellite data, in the area where~~

1 satellite-derived  $\text{SO}_2$  exceeds 0.4 DU. Boxes show 25<sup>th</sup>-percentile, median, and 75<sup>th</sup>-percentile  
2 values, lower whiskers the minimum value and upper whiskers the maximum value.  
3



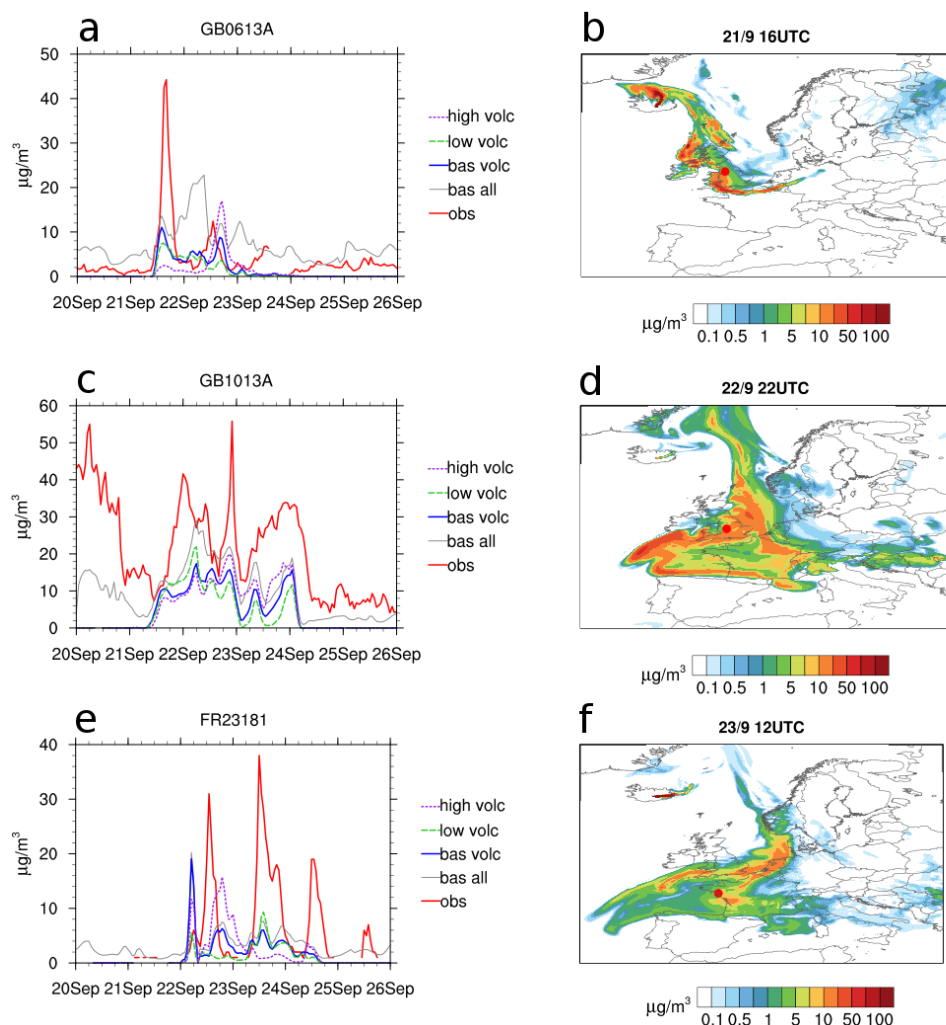
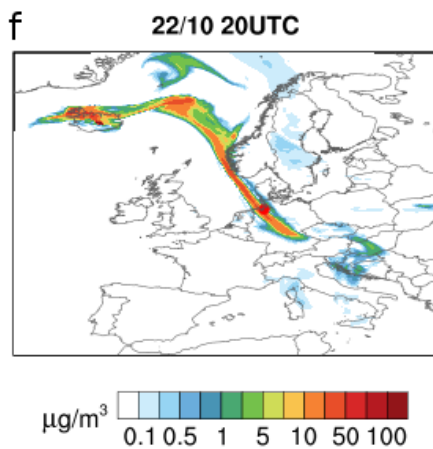
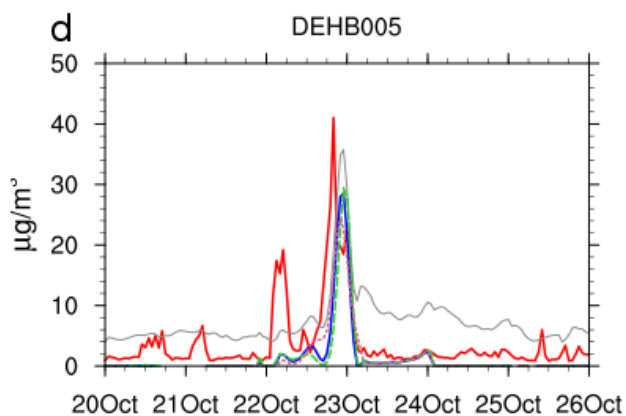
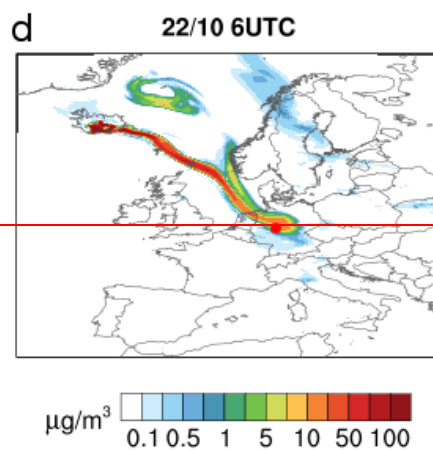
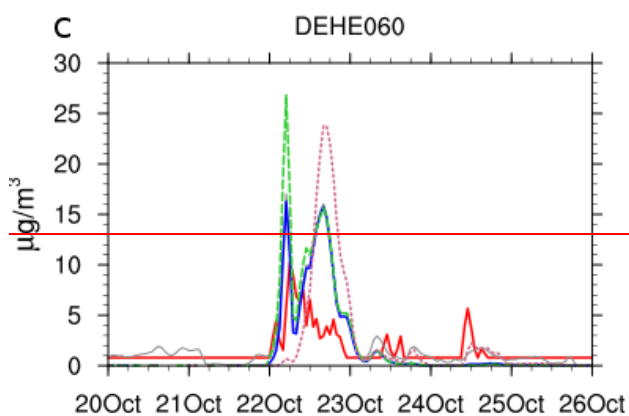
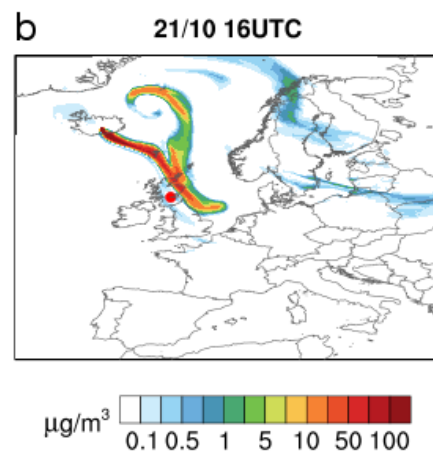
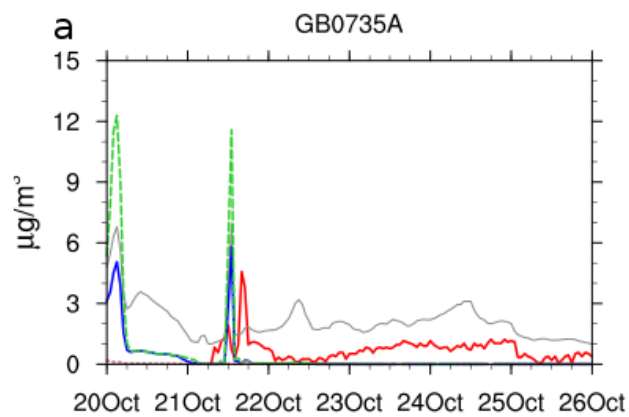


Figure 4. Left: Time Series from 20 to 26 September 2014 for two stations, GB0613A in Manchester and of  $\text{SO}_2$  (top) and  $\text{PM}_{2.5}$  (below) and  $\text{SO}_2$  FR23181 in Saint-Nazaire. The red line shows the measured ground concentrations, the grey line represents the modelled ground concentration with  $\text{et+bas\_hol}$ . By subtracting the ground concentrations from the no\_hol simulation the concentration due to volcanic eruption for the  $\text{et+bas\_hol}$ , low\_hol and high\_hol calculated and are shown in the blue, green and pink line respectively. Right: Ground concentration due to the volcanic eruption from  $\text{et+bas\_hol}$ , corresponding to the blue line in the time series, for the time of the maximum observed concentration. The red dot on the map marks the position of the station.



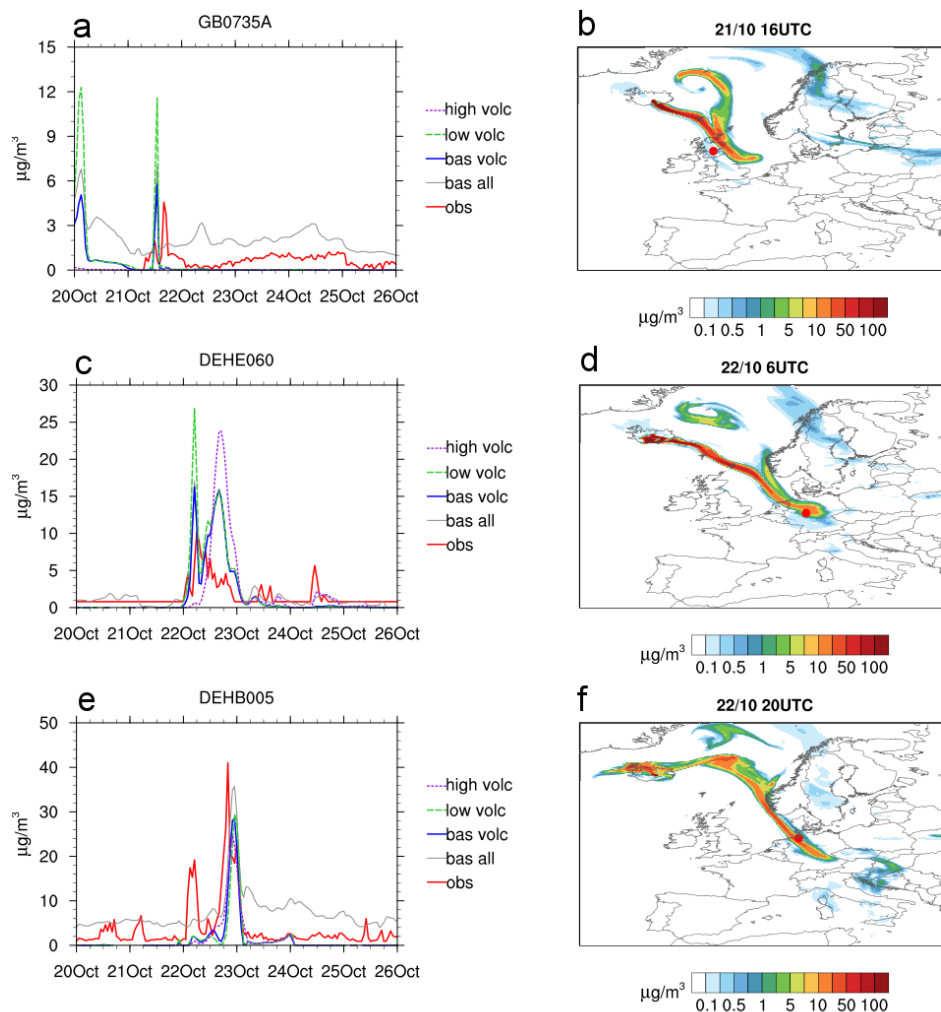


Figure 5. The same as Figure 1, but from 20 to 26 October 2014 for three different stations GB0735A Grangemouth in Scotland, DEHE060 Kellerwald and DEHB005 Bremerhaven in Germany, all SO<sub>2</sub>.

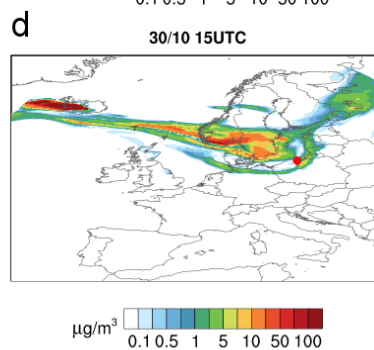
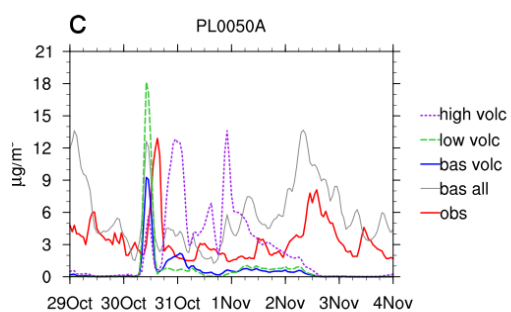
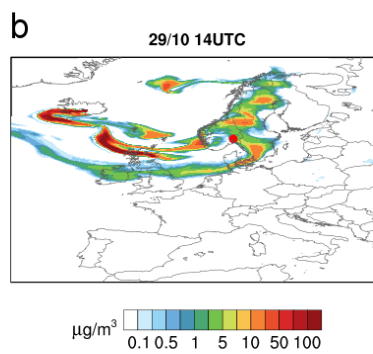
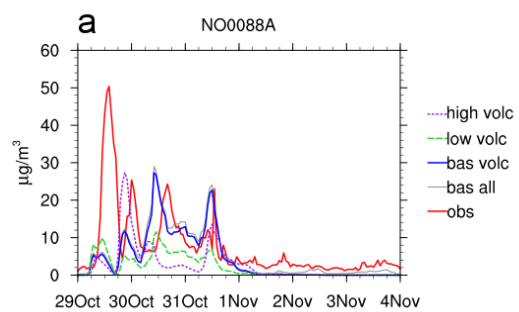
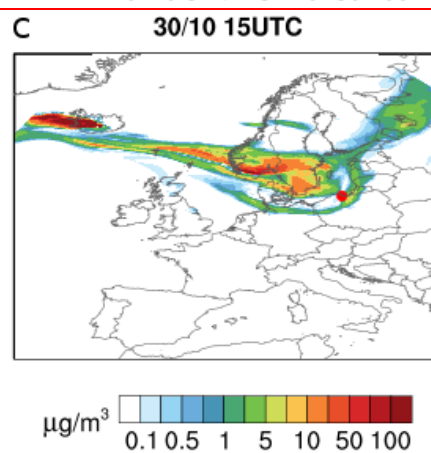
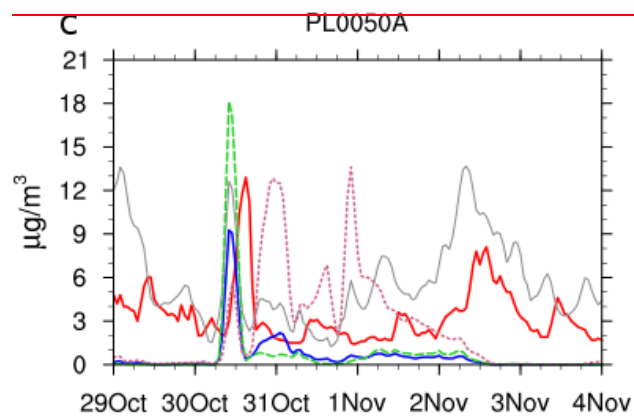
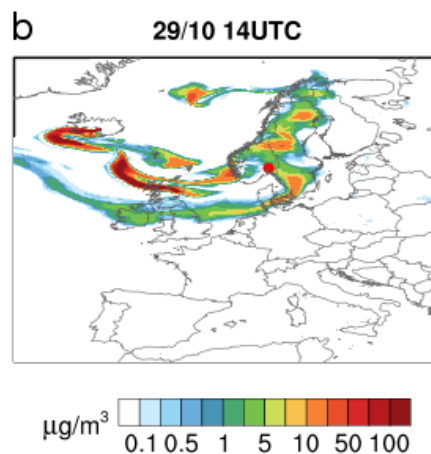
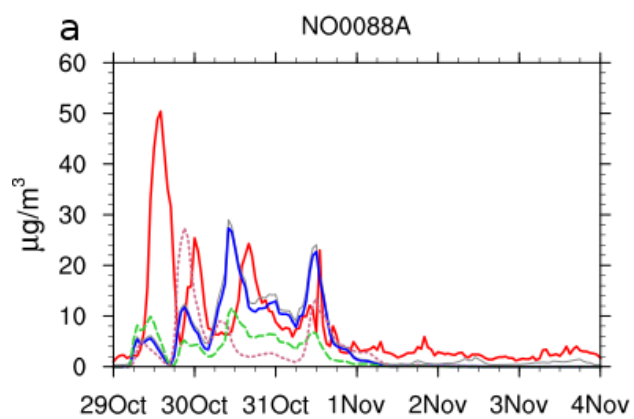




Figure 6: The same as the two previous figures but from 29 October to 4 November 2014 for NO0088A Oslo, Norway and PL0050A in Sopot Poland, both  $\text{SO}_2$ .

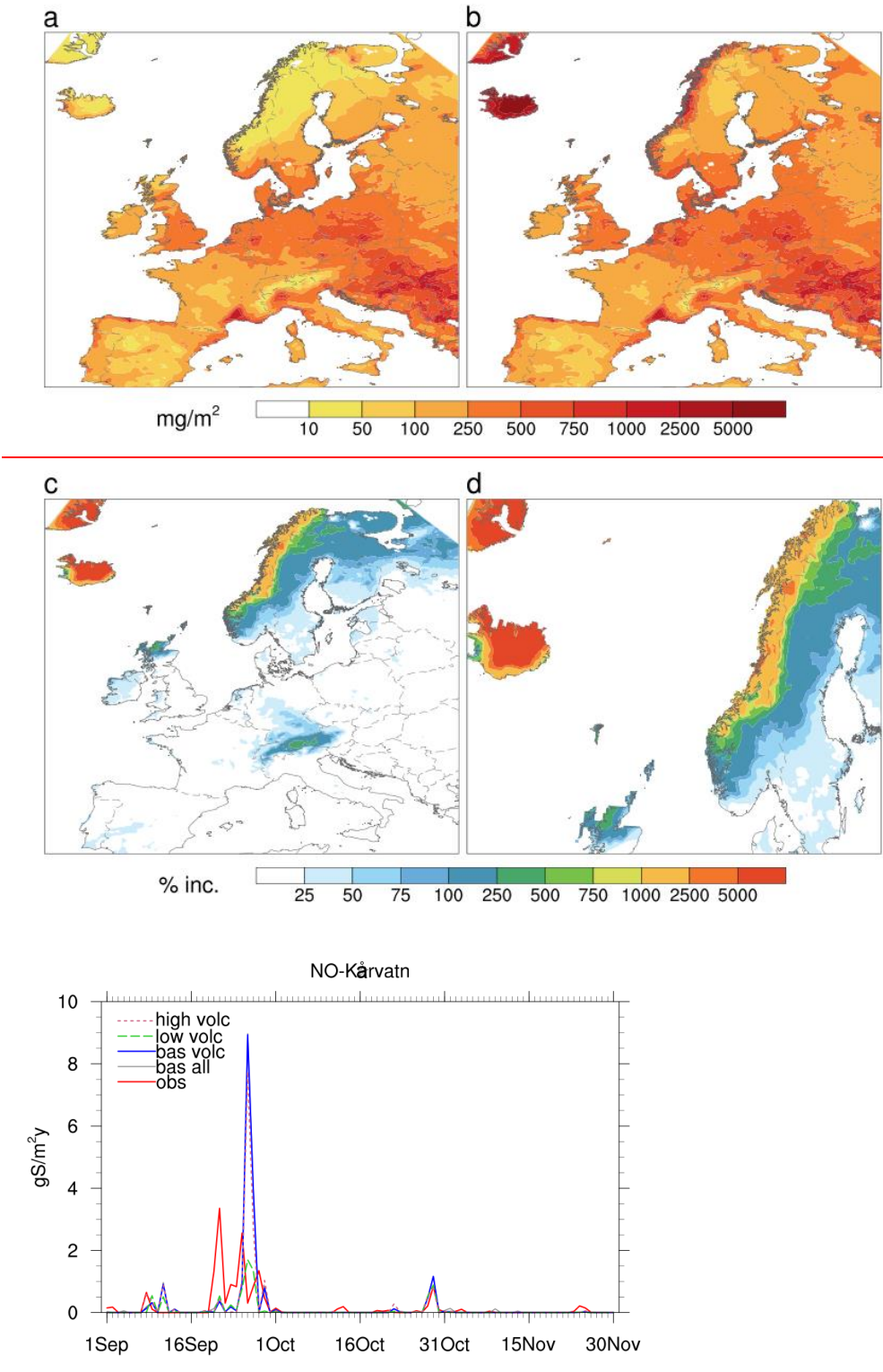


Figure 7. Daily time series of SO<sub>x</sub> deposition from The Kårvatn station in Norway. The lines represent the same as the three plots above.

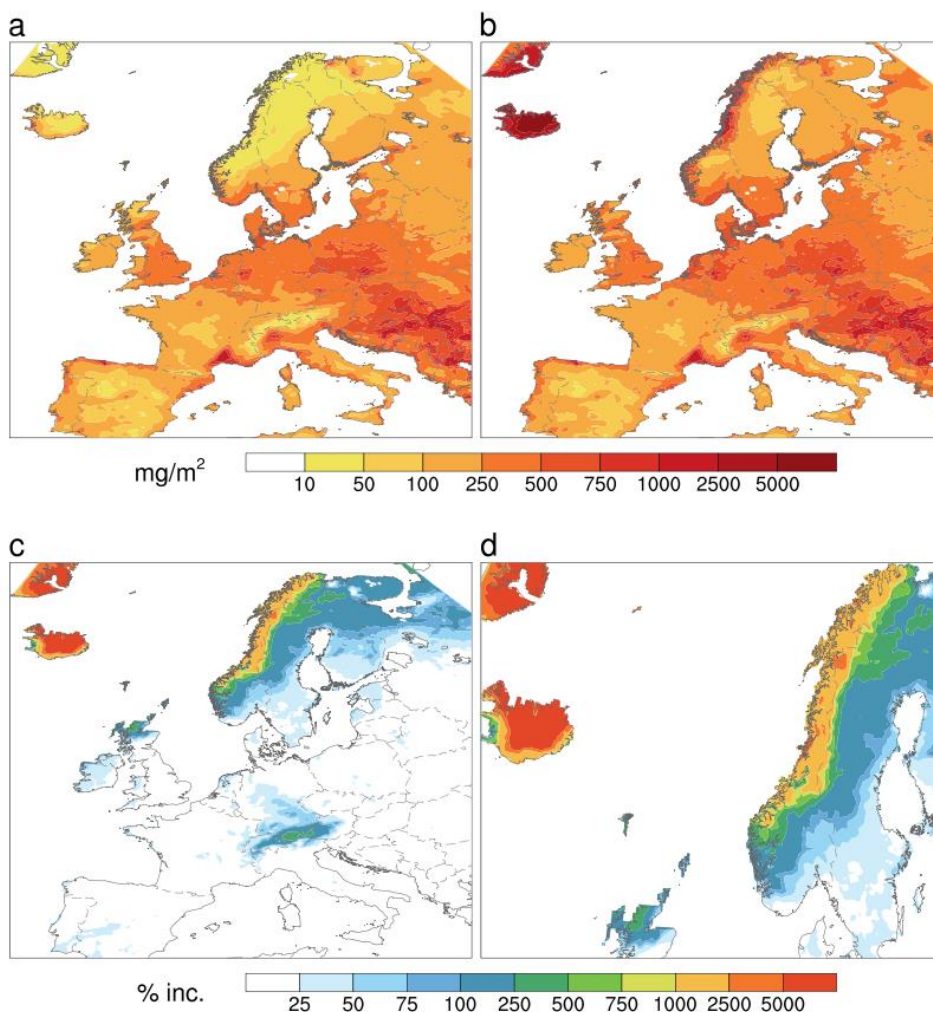


Figure 8. Total deposition of SO<sub>x</sub> (wet and dry) over Europe from September to November for no\_hol (a) and etrbas\_hol (b) simulations and the percent increase due to the Holuhraun emissions (c). d) Shows the same as c) but zoomed into Norway and Northern Europe.

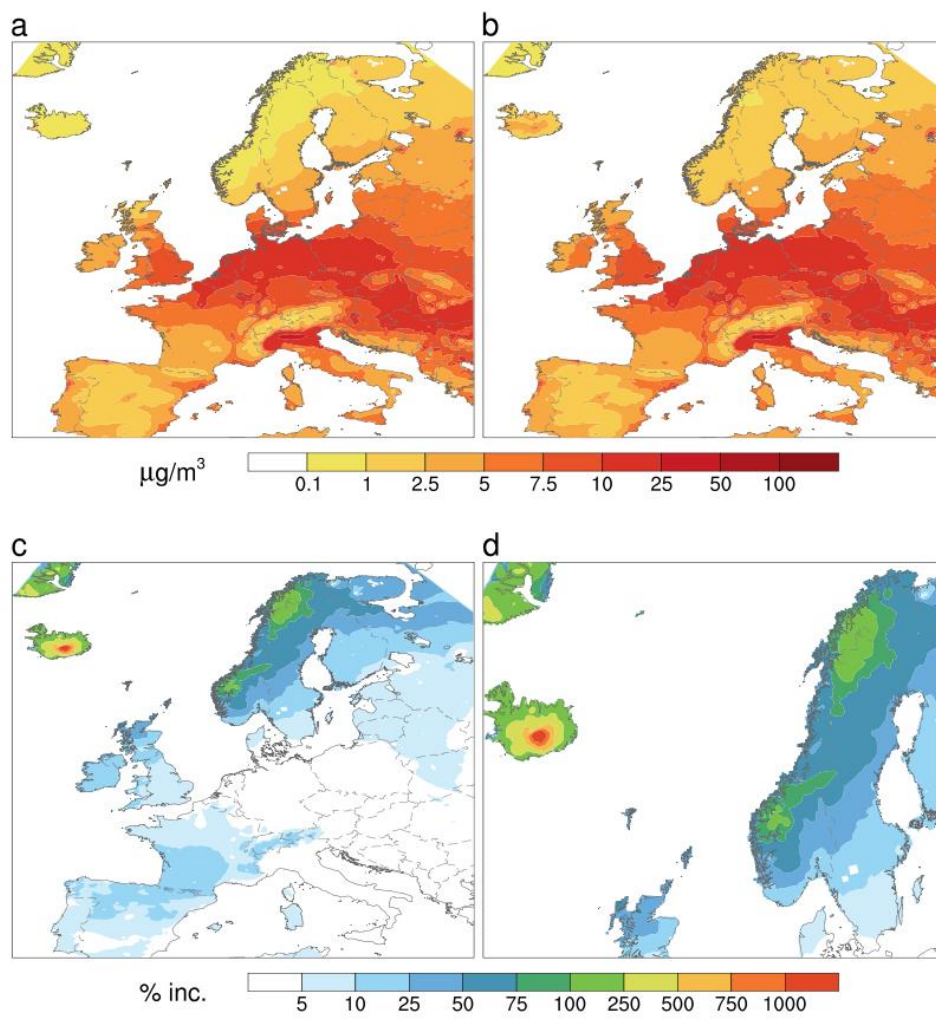


Figure 89. Show the same as Figure 78, but with average PM<sub>2.5</sub> concentration over the three months.

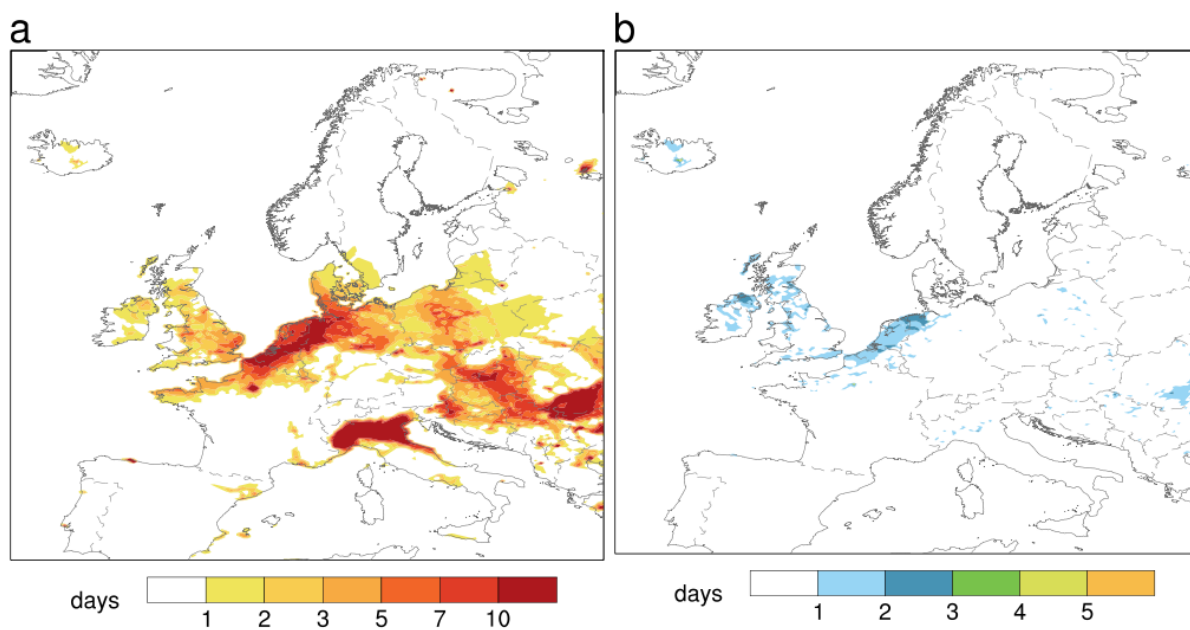


Figure 910. a) Days with exceedances of PM<sub>2.5</sub> over September through November for the `etrlbas_hol` model simulation. b) The increase in days from `no_hol` to `etrlbas_hol`.