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 µg/m3 averaged over an hour of SO_2 from volcanic plumes passing."

You report increases in PM2.5 mass concentrations based on your model simulations. There are plenty of PM2.5 monitoring sites across Europe (many more than for SO2), so you ought make an effort to compare the model simulations to these observations.

$PM_{2.5}$ observations are included in the manuscript for the station in Manchester during the first period when both SO_2 and $PM_{2.5}$ are measured at the station, for the other $PM_{2.5}$ 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 SO2 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 SO_2 and ash plumes. For eruptions where both ash and SO_2 are emitted, 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 SO2 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_2 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.

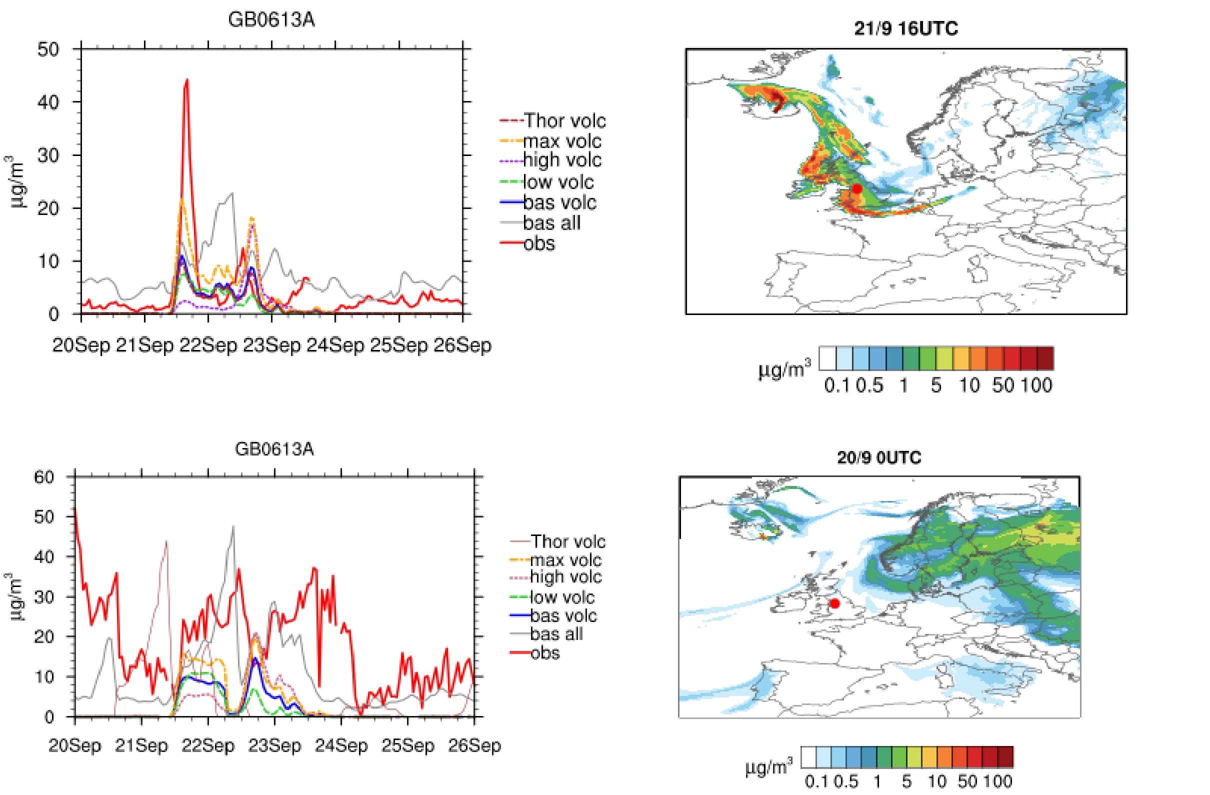

Figure 1. Measured and modelled comcentration at GB0613A station in Manchester, Great Brittain (red dots on the map). The timeseries above show SO_2 concentrations and below for $PM_{2.5}$ for observed (red) and five different model simulations, bas all show all sources for SO_2 and $PM_{2.5}$, while 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 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 SO2 observations for these episodes

Included in the text:

"For the first six day period, between 20 to 26 September, high concentrations of SO_2 were measured over Great Brittain, 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 Brittain, 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_2 for the model data on 7 September, and 37.42 kt SO_2 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 SO2 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_2 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.

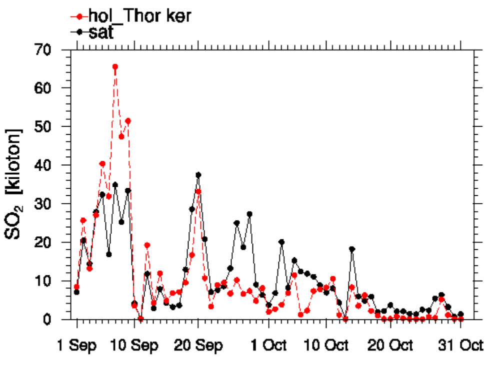

Figure 2. Daily time series of mass burdens from satellite data (black dots) and from model run with Thorarson and Hartley (2015) emission (red dots) with averaging kernel applied.

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 SO2 mass concentrations of about 500 ug/m3 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_2 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.

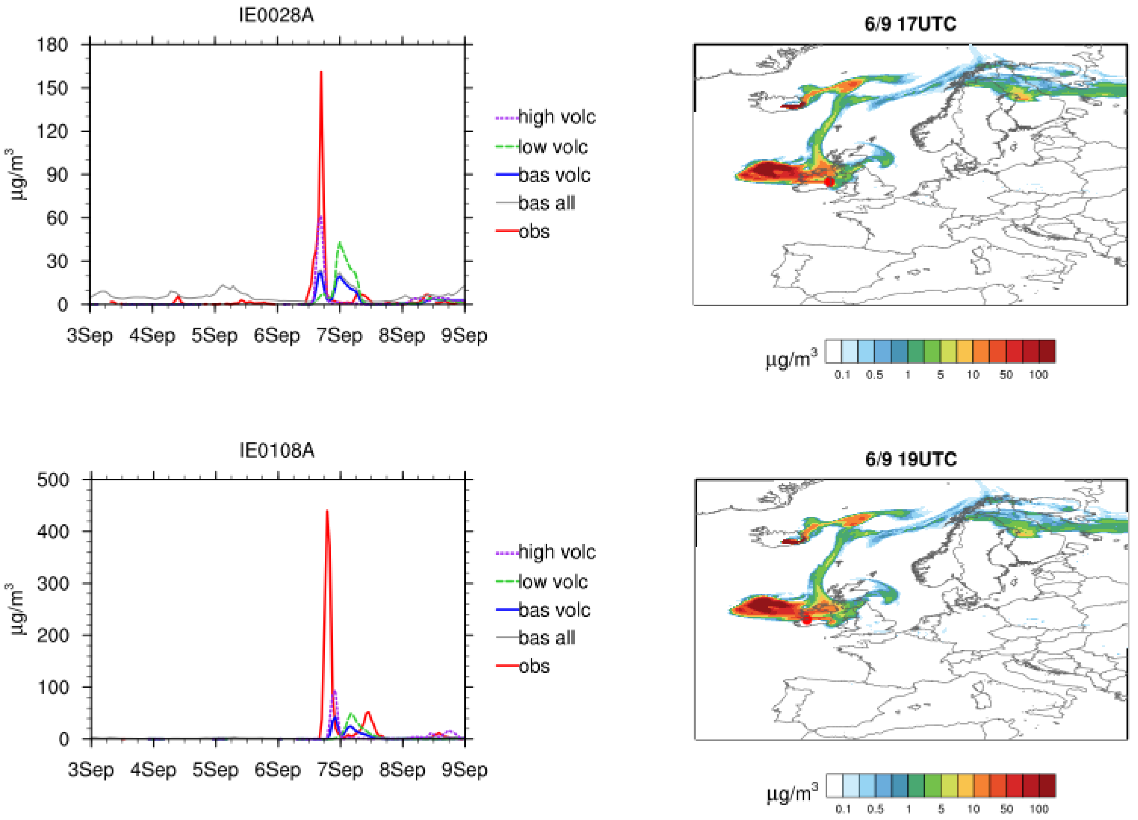

Figure 3. Measured and modelled comcentration at station IE0028A and IE0108A in Ireland (red dots on the map). The timeseries show SO_2 concentrations and for observed (red) and five different model simulations, bas all show all sources for SO_2, while the other volc lines only show values due to the 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 µg/m3 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 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.

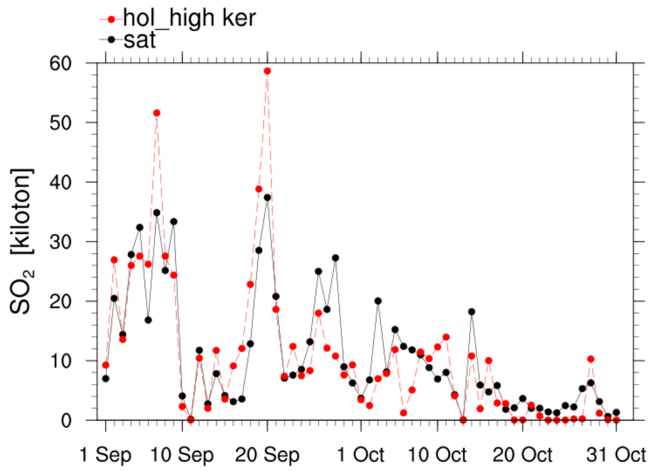

Figure 4. Daily time series of mass burdens from satellite data (black dots) and from model run with emissions released between three and five km, high_hol (red dots) with averaging kernel applied.

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 $PM_{2.5}$ 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 SO2 mass concentrations at the ground compared to the observations).

$PM_{2.5}$ is included in the station comparisons, where a station both measures $PM_{2.5}$ and 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 at 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_2 plume may introduce additional bias between model and satellite VCDs."

Second paragraph: Unless you carry out a sensitivity study changing the SO2 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 SO2' 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_2 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 SO2 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_2 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.