Dear editor,

The authors would like to thank the reviewer and editor for their careful and encouraging comments.

Please find below our response to critical comments, author response are given in italics:

“I certainly think this manuscript should eventually be published, but I still feel that, at times, the writing style is too poor and too qualitative for ACP standards.”

=> We went through the whole document and revised a lot of phrases carefully. A track changed manuscript is provided as part of the response.

“My main quibble is about their sentence saying that pollution increased by up to 50 ug/m^3 (lines 4-6 abstract) this is not true and the authors state that later themselves (lines 1-2 on page 4 of their revised paper in the attached document). They explain in the reply to my comment that they excluded these stations, but to my mind that cannot be a valid reason to state in an abstract that pollution reached up to 50 ug/m^3 because I think people agree that the pollution episodes (> 500 ug/m3) in Ireland on 6 Sep 2014 are linked to Holuhraun.”

=> We have rectified the abstract and included the comparison to the episode in Ireland into the text and graphs into the supplementary material. We had unfortunately not access to all data used by Schmidt et al 2015, and our comparison is thus more limited. It also is not a typical trans-national episode, which was interesting in the other episodes. However, we agree that this episode was very important for the characterisation of the volcanic eruption and that it must be mentioned that concentrations > 500 ug/m3 appeared.

Considering this sentence:

“Surface observations in Europe showed concentration increases up to 50 µg/m3 averaged over an hour of SO2 from volcanic plumes passing.”

the reviewer wrote the following comment: in Ireland SO2 surface concentrations increase to over 500 ug/m3 on 6 Sep 2014. I don't understand this sentence nor is it very specific. The authors should state the period of observation and location. I assume they are not talking about the volcanic pollution episode on 6 Sep 2014 as otherwise the 50 ug/m3 is too low.

=> We have carefully checked for a correct wording around the 6 Sep episode.

Does the terminology 'basic (bas)' in the sentence below refer to the baseline simulation?

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

=> We agree that the naming of the base/reference/bust-guess simulation is difficult. We kept the abbreviation, but added at places in the text, where we thought it useful ‘best guess’ to emphasize the nature of the base run.

In the following text:

“For the first six day period, between 20 to 26 September, high concentrations of SO2 were measured over Great Brittain, and countries to the south. For the second six day period," please add 2014 behind September. The manuscript needs to be checked for such things throughout.

=> We checked where the year 2014 was helpful to add.

With respect to this statement:

"The detailed information will be included in the manuscript. The high SO2 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."

The reviewer had the following comment:

I don't understand why this is a reason to leave out these data. Several stations in Ireland detected high SO2 pollution and backward and forward trajectory analysis shows that it is volcanic pollution.

=> Reviewer is correct, see our comment above, we have added a paragraph to discuss the Ireland episode.

Please rephrase the sentence below that the reviewer deem is neither correct, nor good English:

"Surface observations in Europe showed peak type concentration increases up to 50 µg/m3 averaged over an hour of SO2 concentrations from volcanic plumes passing by and lasting only for a short time."

=> rephrased to

“Surface concentration comparisons presented in this study and in the supplementary material show that the volcanic SO_2 was observed as short singular peaks lasting a few hours or as a sequence of several peaks spread over a few days. Three episodes are picked where transnational transport is documented. “

"The eruption ended in February 2015 and during the 6 months of eruption a total of approximately 11 (± 5) Tg SO2 may have been released (Gislason et al. 2015), and the total lava field from the fissure were 85 km2 with a volume of 1.4 km3 (vedur.is)."

Check wording, is the word area missing from this sentence?

=> reworded to

“and the total lava field from the fissure measured 85 km^2 in area with a lava volume estimated to amount to 1.4 km^3 (vedur.is). “

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~~, with an extensive lava flow onto the Holuhraun plain~~ combined~~. This event was associated with large SO_2 emissions, amounting up to approximately 4.5 times the daily anthropogenic SO_2 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~~kgs^{-1} SO_2 emissions at the Holuhraun plain from September to November~~ at~~, testing three different emission heights. ~~Model results are compared to satellite observations and European surface measurements.~~ The ~~different runs are~~three simulated SO_2 concentrations, weighted with the OMI satellite averaging kernel, ~~the effect~~are found to be within 30% of the ~~weighting are dependent on~~satellite observed SO_2 column burden. Constraining the SO_2 column burden by the satellite data, while using the kernel along with the three simulated height distributions of ~~the sulphur dioxide~~SO_2, we estimate that the median of the daily burdens may have been between 13 and 40 kt in the ~~atmosphere.~~North Atlantic area under investigation. We suggest this to be the uncertainty in the satellite derived burdens of SO_2, mainly due to the unknown vertical distribution of SO_2. Surface observations in Europe outside Iceland showed concentration increases up to ~~50 µg/m3 averaged over an hour of~~ >500 µgm^{-3} SO_2 from volcanic plumes passing. Three well identified episodes~~ are documented~~ , where the plume crossed several

countries, are compared in detail.– to surface measurements. For all the events, the general, timing of the observed concentration peaks compared quite well to the model results. The overall changes into the European SO_2 budget due to the volcanic fissure are estimated. Three -monthly wet deposition of SO_X in the 28 European Union countries, Norway and Switzerland is found to be more than 30 % higher in the model simulation with Holuhraun emission compared to a model simulation with no Holuhraun emission. The largest increases, apart from extreme values on Iceland, are found on the coast of Northern Norway, a region with frequent precipitation during westerly winds. Average On three month average over Europe, SO_2 and $PM_{2.5}$ surface concentrations increase, due to the volcanic emissions, increased by only ten anqdand six percent over Europe, respectively. Although the percent increase of $PM_{2.5}$ concentration is highest over Scandinavia and Scotland, an increase in PM exceedance days is found over Ireland and the already polluted Benelux region, (up to 3 additional days), where aany small increase in pollutionparticulate matter concentration leads to an increase in exceedances days.

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_2) emitted into the atmosphere (Sigmundsson et al. 2015). Thordarson and Hartley (2015) estimated SO_2 emissions from the magma at Holuhraun to be aroundrange between 30 kt/d toktd^{-1} and 120 kt/dktd^{-1} 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/dktd^{-1}) 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/dktd^{-1}, while the 2013 estimate is 9.8 kt/dktd^{-1} (EMEP, 2015). The eruption ended in February 2015 and during the 6 months of eruption a total of approximately 11 (± 5) Tg SO_2

may have been released (Gislason et al. 2015), and the total lava field from the fissure ~~were~~measured 85 km^2 in area with a lava volume ~~of~~estimated to amount to 1.4 km^3 (vedur.is). It is of interest to investigate the impact of these volcanic emissions on ~~current~~ SO_2 levels in Europe in 2014. In the last decades, measures have been taken to reduce SO_2 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~~Tørseth et al., 2012). The impact of volcanic eruptions with SO_2 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_2. 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_2 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 5 km (vedur.is). Ground level concentrations exceeded the Icelandic hourly average health limit of 350 ~~$\mu g/m^3$~~$\mu g m^{-3}$ over large parts of Iceland (Gislason et al. 2015). The World Health Organization (WHO) has a 10 minute limit of 500 ~~$\mu g/m^3$~~$\mu g m^{-3}$ and a 24-hour limit of 20 ~~$\mu g/m^3$~~$\mu g m^{-3}$. High hourly mean surface concentrations of SO_2 were measured in Ireland (524.2 ~~$\mu g/m^3$~~$\mu g m^{-3}$), but then also in Austria (247.0 ~~$\mu g/m^3$~~$\mu g m^{-3}$) and Finland (180 ~~$\mu g/m^3$~~$\mu g m^{-3}$) (Schmidt et al. 2015, Ialango et al. 2015).

A climate impact of high SO_2 emissions may be suspected, such as a cooling of climate due to an increase in aerosol ~~loadings~~burdens. Gettelman et al. (2015) using a global climate model found a small increase in cloud albedo due to the Holuhraun emissions resulting in -0.21 Wm^{-2} difference in radiative flux at the top of the atmosphere. If the event had happened earlier in the summer a larger radiative effect could be expected (-7.4 Wm^{-2}). Understanding the atmospheric sulphur budget associated to such events is thus of great interest also for climate science. Unlike the two previous big eruptions in Iceland, Eyjafjallajökull in 2010 and Grímsvötn in 2011, this eruption did not emit important amounts of ash. However, uncertainties in volcanic source estimates, time varying emissions from a volcano type of 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 will occur because of density differences and different eruption heights (Moxnes et al., 2014). Proven capability of modelling the transport of a volcanic plume can be useful for judging future eruption scenarios where SO_2 or ash can cause a problem.

The Holuhraun eruption is worth being analysed for several gas and aerosol transport and transformation processes, this study will mainly focus on simulated air quality effects and the perturbed sulphur budget due to the volcanic SO_2 emissions during the first three months of the eruption. Several stations in Europe reported high concentrations of SO_2 during this time and case studies are chosen to evaluate simulated plume development over Europe. The transport is modelled with the EMEP/ MSC-W chemical transport model, one of the important models used for air quality policy support in Europe during the last 30 years (Simpson et al. 2012). The first two months of the eruption are well covered by satellite observations. Both station and satellite data are compared to model results to understand the amplitude and magnitude of the sulphur budget perturbation. The effect of the injection height on the model results is studied by sensitivity simulations. Finally the perturbed European sulphur budget, is documented and discussed to investigate the impact of increased SO_2 emission from a Icelandic volcano on European pollution levels.

2 Methods

2.1 Model description

The model simulations of the transport of the SO_2 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/ MSC-W model is described in Simpson et al. (2012). SO_2 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^o (longitude) x 0.125^o (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^o west to 45^o east and 30^o to 76^o north). Iceland is in the upper 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/ MSC-W 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/skg s^{-1} SO_2 (65 kt/dkt d^{-1}) for the entire simulation from the total 2.0 ± 0.6 Tg SO_2 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. 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 runbest guess, base case simulation, 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. 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 3km3 km and 5 km called high_hol. To derive the impact purely due to the emissions from Holuhraun, a simulation with no Holuhraun emissions is performed, called no_hol. Sensitivity runs with an almost doubled constant emission rate of 1400 kg/skg s^{-1}, 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_2 emissions in the model are described in Kuenen et al. (2014). There is a yearly total SO_2 emission of 13.2 ~~Tg/a~~Tga^{-1} 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~~Tga^{-1} SO_2 in yearly ship emissions over the oceans. Over the continents the yearly emissions are 5.08 ~~Tg/a~~Tga^{-1} SO_2 for the 28 EU countries, and 5.53 ~~Tg/a~~Tga^{-1} SO_2 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_2 concentrations are often too low to be observable, but increases in SO_2 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_2 molecules varies with altitude (generally decreasing towards the ground level) and therefore the algorithms use an assumed height distribution for estimating the integrated SO_2 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_2 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). As found in Schmidt et al. (2015), this is too high for the ~~Bardarbunga~~Bárðarbunga eruption. Therefore, the- retrieved SO_2 column densities may 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^o west to 15^o east and 45^o to 70^o north (red boxes in ~~Figure~~Fig. 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 post processing. 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_2 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_2. Standard deviation for the satellite data is calculated over an apparently SO_2 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~~above 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_2 VCD in the grid cells above the threshold. Finally we convert here and there in the manuscript DU to mass burdens to facilitate comparison to models and mass budgets. One DU is 2.69 10^{20} molecules per square metre, which corresponds to a column ~~loading~~burden of 28.62 milligrams SO_2 per square meter (~~mg/m^2~~mgm^{-2}).

Data of SO_2 and $PM_{2.5}$ 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_2), and a second one obtained from EEA as so called up-to-date (UTD) air quality data base, state spring 2016. 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, 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 only some outstanding episodes with high concentrations of SO_2 and documented trans-national transport of a volcanic plume are analysed. For the first six day period between ~~20th~~20 and ~~26th~~26 September 2014, high concentrations of SO_2 were measured over Great Britain and countries further to the south. For the second six day period, a month later (~~20th~~20 to ~~26th~~26 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 were 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_2 location and column density. ~~Figure~~Fig. 1a shows as an example the VCD from the OMI satellite overpasses on 24 September 2014. Fig. 1b and Fig. 1c show the modelled and the ~~adjusted~~kernel weighted VCD from the ~~basic run~~base simulation (bas_hol). The observed satellite SO_2 cloud and the model simulated SO_2 cloud show similar shape and location. The ~~adjusted~~kernel weighted 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, where there is less SO_2 in our case, with emissions and transport happening in the lower part of the troposphere. The reduced kernel weighted column densities are ~~more comparable~~closer to the column densities observed by the satellite. There are however some spatial 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~~burdens. Fig. 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 valid 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~~cut-off 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~~burden is above the model value, ~~not including~~ignoring the days where the OMI zoom mode ~~minimizes the~~ is responsible for a small coverage. The average satellite derived SO_2 mass burden ~~adjusted to the~~assuming a 7 km reference height ~~for satellite data are 11.17 kt SO_2 for satellite and~~is 11.2 kt, while the kernel weighted model burden in bas_hol is 8.~~72~~7 kt SO_2 ~~for the model.~~. The highest values are found at the beginning of the period, 42.~~11~~1 kt SO_2 ~~for the model data~~ on 7 September, for the model, and 37.~~42~~4 kt SO_2, on 20 September for the satellite ~~data.~~. Taking into account the area in which the satellite observed SO_2 is found above detection limit, the satellite average column loadings are calculated to reach 70 mgm^{-2} for September. Also the peaks in the middle of October, visible in Fig 2b, exhibit a satellite average column loading of 62 mgm^{-2}.

The daily values of SO_2 mass burden are decreasing over time, especially during October. There is also a positive bias of the model against the satellite in the end of October. At the same time the satellite coverage is decreasing~~.~~ along with an increasing solar zenith angle. To further investigate whether ~~the increasing solar zenith angle~~this is responsible for the general

Formatert: Senket

decrease in mass burdens and the increasing bias of the simulated versus observed VCDs, a new aggregation domain further south is used. All 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~~burdens 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_2 from Holuhraun first needs to be transported south ~~to~~before it can be detected. The plume is transported that far south four times over the two-month period as the peaks in column ~~load~~burden values in Fig. 2c show. In this southerly area the daily accumulated mass burdens are similar in September and in October, supporting the ~~idea~~hypothesis that the apparent 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_2 above detection limit, the satellite average column loads are calculated to around 70 mg/m^2 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^2.~~

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_2 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~~3. Note that the mass burdens are accumulated in the same area in the North Atlantic, where at least 0.4 DU SO_2 was observed by OMI. Looking at the original model data, the model simulation with emissions in the lowest kilometre (low_hol) has the highest daily mass burden values~~,~~ (median: 25.4 kt), followed by the best guess simulation, bas_hol (median: 22.5 kt), while the run with the emission highest in the atmosphere (high_hol) exhibits ~~a lower~~the smallest mass burden ~~than the two other.~~(median: 15.9 kt). 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_2 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_2 at high altitudes.~~

~~Comparing the satellite data to the kernel weighted model data; the satellite 75^{th} percentile is higher~~The kernel weighted model data represent what can be directly compared to the satellite data. As shown in Fig.3, and illustrated already in Fig. 1, the kernel weighted model column burden values are much smaller than the ~~model 75^{th} percentile.~~ original ones, because the SO_2 plume was simulated to be much below 7 km altitude. The impact of the kernel weighting is quite different for the three model simulations. After the averaging kernel has been applied to the model data, the high_hol model simulation exhibits the highest daily burdens compared to the other two model simulations. The median for the bas_hol, low_hol and high_hol daily mass burden are 7.~~38~~4 kt, 4.~~43~~4 kt and 8.~~34~~4 kt respectively, ~~for~~while the satellite ~~the~~ mass burden median value is 7.~~03~~0 kt. ~~The~~High burdens retrieved from satellite data ~~therefore have higher maximum values~~, and high kernel weighted model burdens reflect that ~~results in the higher average values~~volcanic SO_2 is present at high concentrations and ~~the 75^{th} percentile, most~~/or at high altitudes.

Analysis of the ~~satellite~~distribution of daily mass ~~burden values are however around the model data for the bas_run. From all the model simulations,~~burdens allows investigating how many days with very high burdens were present. Comparing the satellite data to the kernel weighted model data we find that the satellite 75^{th} percentile is larger than any of the model simulation's 75^{th} percentiles. The satellite data contain some high daily burden values that result in a higher average burden and a higher 75^{th} percentile. From our three model simulations, testing different emission heights, the best-guess bas_run ~~is~~has got the most similar distribution of daily burdens compared to the satellite data over the first two months.

3.2 Surface concentrations

SO_2 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~~A particular episode with very high surface concentrations of up to 500 µgm^{-3} SO_2 in Ireland in the beginning of September was studied by Schmidt et al. (2015). However, just very few Irish station data were in the data extract we obtained from the EEA for this episode and we decided to document the comparison for this episode in the supplementary material.

The comparison supports, however, that our emission flux indeed might have been too small in the first days of September 2014.

Fig. 4 shows hourly time series for two stations over Great Britain and France from 20 September to 26 September. ~~Concentrations~~A maximum concentration of 44.~~25 µg/m^3~~3 µgm^-3 SO_2 ~~concentrations were~~was measured 21 September 16 UTC at 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. However, the ~~model~~simulated concentration field shows areas south of the station nearby Manchester, where the volcanic SO_2 concentrations ~~only due to the volcanic eruption~~ are around 50 ~~µg/m^3~~µgm^-3. Interestingly, the agreement of the model derived volcanic SO_2 time series is better in agreement with measurements than the total simulated SO_2 concentration (grey curve), indicating that the model may not resolve SO_2 transport from nearby pollution sources and that the station for these days is rather representative of long range transported volcanic SO_2. Observed $PM_{2.5}$ ~~concentration~~concentrations at the station ~~shows~~show, that over the period, the highest concentration (52.1 ~~µg/m^3~~µgm^-3) – probably anthropogenic - is measured at the start of the period, ~~before~~when the model did not simulate any volcanic sulphur contribution~~ is simulated by the model~~. The next day, the plume has moved further south over France, the station is situated 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 ~~µg/m^3~~µgm^-3) measured ~~12 UTC~~on the 23 September at 12UTC. All the three model simulations have the peak concentrations earlier than the observed ones, 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 SO_2 surface concentrations during this time.

~~Figure~~Fig. 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 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 on 21 October for the two models is a few hours early and the modelled concentrations higher than ~~the~~ observed (6.09 ~~µg/m^3 – observed,~~µgm^-3), especially ~~for~~in 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 SO_2 is measured at stations in Germany. ~~Figure~~Fig. 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 ~~µg/m^3~~µgm^{-3}) is short in duration, the peak lasts for one day at Kellerwald with an observed maximum of 10.2 ~~µg/m^3~~µgm^{-3}. The map shows that the plume is narrow for all three stations and the local spatial gradient ~~between where there~~ is ~~no Holuhraun contribution and the maximum concentration is strong~~large.

A third plume is illustrated in Fig. 6 over Northern Europe, occurring from the end of October to the beginning of November. ~~Figure~~Fig. 6a shows the measured 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 ~~µg/m^3~~µgm^{-3}). The models runs show contribution from Holuhraun 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~~, With the~~. The bas_hol simulation ~~has~~exhibits the most ~~comparable concentrations~~similar concentration evolution among the three model experiments.

~~Figure~~Fig. 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~~in Norway. There are high levels, both observed and modelled during the last part of September. The model ~~exhibite high values~~exhibits a clear peak value on 27 September, while the ~~observed~~observations record deposition ~~is~~ spread out over several days. Summed over the whole period, the ~~observation has~~observed deposition amounts to 15.9 ~~gS/m^{2}y~~gSm^{-2}y^{-1} while the ~~bas~~ model simulated ~~19.98 gS/m^{2}y.~~20.0 gSm^{-2}y^{-1}. Comparisons at other stations in Norway ~~also~~ show ~~the same~~similar results (~~appendix~~supplementary information).

~~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_2 cloud south over Great Britain and France. Compared to the other two episodes, the SO_2 surface concentration due to Holuhraun are higher over a larger area during this episode. The difficulty of the model~~

~~to simulate the SO_2 transport correctly depends on the uncertainty in the emission term, the meteorology fields, the chemical reactions and deposition. Overall the comparison to observations shows, taking into account satellite and station data, that the bas_hol model simulation matches best with the observed satellite column burdens, their time evolution and for some stations with the magnitude and timing of the observed peaks.~~

3.3 Effects of the eruption on European pollution

The above results ~~above~~ show that, ~~although~~despite the Holuhraun eruption ~~released~~releasing large amounts of SO_2, the stations in Europe often measured ~~the~~an increase in SO_2 concentration only as short peaks (Grislason et al. 2015, Schmidt et al. 2015). The model makes it possible to ~~find a more~~ investigate the general ~~view of the~~ impact ~~in the~~on European air quality ~~due to the~~by Holuhraun volcanic emissions. Table 2 summarizes ~~the model results~~ characteristic SO_X budget terms and surface concentrations for ~~Europe. Grid cells covered by the~~ the European continental land area in the countries mentioned ~~are used for calculating the results shown~~ in ~~the~~ table~~. The emission (from anthropogenic sources), concentration~~ 2. Concentration and deposition over the oceans are not included. ~~Since a large part of the deposition and concentration increase occurs downwind and to~~To isolate the effect of the Holuhraun eruption on Iceland itself ~~close to the emission point,~~ the deposition and concentrations over Iceland are given in brackets.

The table shows, that the Holuhraun emission ~~estimate used~~flux in ~~this~~the study ~~releases~~period corresponds to over 4.5 times the anthropogenic emission from the 31 European countries considered here (not including ship emissions). The anthropogenic emissions from Iceland are only 18 kilotons, the SO_2 emissions from Iceland increase by more than 300 times.

Over the three months, there is 1.32 times more SO_X wet deposition for the ~~basic~~base run with Holuhraun emission than the ~~MACC~~ reference simulation with no Holuhraun emission~~. Wet~~ (no_hol). Table 2 shows that wet deposition over Europe is quite dependent on the emission height. The simulation with the emission highest in the atmosphere (high_hol) ~~has~~exhibits the highest ~~contribution to the rest of~~wet deposition in Europe. For dry deposition, ~~the~~a ten percent increase over Europe is ~~about the same~~found for all the three model simulations with Holuhraun emissions. Close to the source, over Iceland, the deposition levels are very dependent on ~~the emissions~~emission height, ~~especially for~~ dry deposition ranging from 8 to 409 kilotons.

~~Figure~~Fig. 8 shows the total deposition over Europe for the standard MACC model simulation with no Holuhraun emission (no_hol), the ~~basic~~base model simulation (bas_hol), and the percent increase between these two model runs. Areas that experience the highest percent increase are also areas that have low levels in the model simulation with no emission at Holuhraun. Due to the Holuhraun emissions Iceland has the highest SO_X deposition in Europe, and the coast of northern Norway shows depositions on the same level as the more polluted ~~eastern~~Eastern Europe. Even though the previous section indicated that the model has higher wet deposition levels in northern Norway than observed, it also showed that it is very likely that the observed increases in SO_x deposition levels are due to the Holuhraun emissions.

The averaged SO_2 surface concentration over Europe is under normal conditions higher than over Iceland, the volcanic emission caused the concentration level over Iceland to increase by a factor of 177 (for the low_hol simulation). Over the rest of Europe the increase is ~~around~~about the same for all three Holuhraun simulations, even though the time series showed that the different simulations had peaks arriving at often different times. ~~On~~Vertical mixing, on average, ~~however, vertical mixing has levelled~~levels off initial differences in emission height ~~when~~for volcanic plumes ~~arrive~~arriving in Europe.

The small increases in $PM_{2.5}$ concentrations over Europe, as shown in table 2, are due to increased sulphate production from volcanic SO_2. However, $PM_{2.5}$ is a collection of all aerosols under 2.5 µm, ~~therefore~~and the ~~increased~~volcanic sulphate is changing total aerosol mass therefore relatively little. The table shows that Iceland has a lower average concentration than the rest of Europe for all the four runs, even though Iceland is the ~~origin to~~source for the increase in aerosol pollution levels. The high_hol model simulation has a higher increase in $PM_{2.5}$ concentration over Europe than the two other simulations. ~~The~~ By contrast, the low_hol simulation finds highest sulphate and SO_x deposition on Iceland itself, and possibly over the nearby ocean, ~~that~~which will lead to a ~~lower~~smaller contribution to pollution levels over the rest of Europe.

The distribution of $PM_{2.5}$ from the no_hol and bas_hol simulation, plotted in ~~Figure~~Fig. 9, shows the same polluted and clean areas as in Fig. 8, although the increase is lower. Over north-west Norway and northern Norway the increase is over 100 percent, ~~Figure~~Fig. 9b shows that although the percentage increase is high, the $PM_{2.5}$ concentrations in these areas are still among the least polluted in Europe. The high deposition levels in this region indicate that some of the $PM_{2.5}$ is scavenged out.

WHO recommends a 24 hourly average mean concentration level of 25 ~~$\mu g/m^3$~~μgm^{-3} for $PM_{2.5}$ not to be exceeded over three days over a year (WHO, 2005). ~~Figure~~Fig. 10a shows that over the Benelux region, northern Germany and northern Italy this limit value is exceeded by up to ten days during the three months studied. As the previous plot showed, these are regions with high average $PM_{2.5}$ concentrations. Because the daily concentrations are already high, any increase in days in the model bas_hol simulation due to the Holuhraun emissions is also occurring in these regions, and the areas with the highest percent increase ~~does~~do not experience any days over the limit. The Figure also shows that Northern Ireland experienced up to two exceedance days due to the volcanic eruption.

4 Discussion

The bias and variances between the ~~satellite~~ model data and the satellite observations can be due to several factors. a) The model ~~emissions~~emission 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~~for which the column mass ~~are constructed~~burdens have been computed depend on the ~~threshold~~ VCD ~~value~~detection limit and the actual satellite data, so the ~~values in the~~retrieved model burdens depend on the position of the identified and observed SO_2 cloud. If ~~the~~a simulated plume is displaced into an area where the satellite does not show any ~~useful~~valid signal or no signal above detection limit, then this part of the model plume is ignored and may lead to underestimates ~~of~~by the model. c) The presence of clouds can increase the uncertainty of the satellite retrieval. ~~d)~~d) The fluctuating real height of the SO_2 plume may introduce additional bias between model and satellite VCDs. Schmidt et al. (2015) ~~presents~~presented IASI (Infrared Atmospheric Sounding Interferometer) plume heights for the Bárðarbunga SO_2 plume 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.~~

Schmidt et al. (2015) ~~presents~~presented a comparison between model, satellite and ground observations for September. Mass burdens from OMI ~~are~~were derived using observed plume heights from the IASI instrument on the MetOp satellite. The model Both satellite data sets were compared with the model NAME (Numerical Atmospheric-dispersion Modelling Environment), a Lagrangian model, iswhich was run for September, with sensitivity runs testing both emission height and emission flux. Comparing with the two satellite data sets, the model The model simulation with a plume height of 3 km and doubled emission flux (~1400 kg/s) matcheskgs^{-1}) matched well with the OMI satellite data for the first days, while for the rest of September the modelanother simulation with emissionmatched better, where emissions were similar to the constant emission term used here matches betterin our study (~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 methodskgs^{-1}). Their and our study 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, the satellite data exhibit higher values than a model using an emission rate of September, but overall the height of the plume is more important for the satellite comparison.

700-750 kgs^{-1} SO_2. Our Holuhraun emission term in the three model simulations is constant throughout the simulations both with respect to the respective three emission heightheights and emission flux. (see table 1). Maximum fluxes of 1300 kg/skgs^{-1} were reported by Barsotti (2014), and). Gislason et al. (2015) estimated a 2.5 times theabove average emission term during the first two and a half weeks of the eruption. TheOur assumption of a constant emission term is thus certainly a simplification.

However, here we suggest that overall understanding of the height of the plume is as important to achieve model agreement with the satellite data as emission intensity variations. The emission height is also variable, dependent on initial volcanic eruption characteristics and meteorological conditions like wind speed and stratification (Oberhuber et al. 1998). Table 1 contains the original mass burdens and the kernel weighted mass burdens as described in section 3.1. It also contains a scaled burden estimate, assuming that each of the three simulations should be corrected for bias against the satellite derived burden. This scaling assumes that in each model simulation the height distribution may be correct. The resulting mass burdens from the three simulations differ by 60%, computed as standard deviation. This may be seen as an uncertainty estimate associated with our limited knowledge of the real height of emission and dispersion of the SO_2 plume from Bárðarbunga.

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A better source estimate for the eruption is beyond the scope of this study~~;~~, however the fluctuations both in flux magnitude and emission height can explain some of the differences between satellite observed and simulated concentrations, especially in the first days of September~~ in the satellite comparison~~.

Surface concentration comparisons presented in this study and in the supplementary material show that the volcanic SO_2 was observed as short singular peaks lasting ~~from~~ a few hours ~~to~~ or as a sequence of several peaks spread over a ~~short set of~~few days. Three episodes are picked where transnational transport is documented. The biggest difference ~~for~~ between simulated and measured concentrations is found for the first of the three studied plumes ~~is for the first one~~ during 20 to 26 September ~~for the~~in Manchester (station GB0613A~~) in~~), Great Britain and ~~the~~ Saint-Naizaire (station ~~(~~FR23181~~) in~~), France, with up to a factor of four differences~~ between simulated and measured concentrations~~. Both the measured and simulated concentrations during the September event were higher than the two later events, pointing to a ~~different~~more efficient transport of SO_2 in ~~the first~~this event~~, and not only higher emissions.~~. Higher emission fluxes up to a factor of 2 are ~~also not~~ supported by the satellite comparison ~~over these~~ on some of the days ~~either~~in the end of September. 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 surface concentrations ~~and burdens~~ well below those observed. Station data comparisons presented in Schmidt et al. (2015) for these days ~~show the same results~~are similar, indicating that ~~the~~ models ~~and meteorology had~~have difficulties representing this period.

The discrepancies found between ~~the~~ model and observations~~, especially for the station data show~~ suggest that the ~~values~~ volcanic SO_X budget terms and average European surface concentrations presented in Table 2 contain error. Especially the model surface concentrations ~~are~~seem to be low compared to observations; however the map plots in Fig. 4, 5, 6 show, that sometimes modelled concentrations nearby the stations reached the observed levels. The area averaged ~~concentrations~~volcanic concentration contribution presented in table 2 may therefore be close to ~~the real concentration increase.~~reality. A more thorough study ~~of longer time series~~ with ~~deposition and concentration trends~~a completed quality controlled data set is needed to estimate better the increase in SO_2 concentrations due to the eruption at the stations in the 2014 volcanic eruption episode.

Transport from an Icelandic volcano to Europe is caused by northerly and north-westerly winds. For the first plume 20 to 26 September, where the model shows low concentrations compared to the observations, there had been southerly winds for some time before strong northerly winds transported the SO_2 cloud southward over Great Britain and France. Compared to the other two episodes, the SO_2 surface concentration due to Holuhraun are higher over a larger area during this episode. The difficulty of the model to simulate the SO_2 transport correctly is connected to the uncertainty in the emission term, the meteorology fields, the chemical reactions and deposition. Overall the comparison to observations shows, that our best guess bas_hol model simulation matches best with the observed satellite column burdens, their time evolution and for some stations with the magnitude and timing of the observed surface concentration peaks.

The results in this study show that the sulphur ~~depositions~~deposition from September to November over Northern Norway ~~were~~was at the same ~~levels~~level as found in the most polluted regions in Europe. ~~Emission~~The emission ceilings aim, set by the Gothenburg Protocol, was to reduce the SO_x emissions by 63 % by 2010 compared to the 1990 levels in the European area of the convention of long-range transport of air pollutants (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 observed averages since 1980. Monthly observed values at this station during the 2011 Grimsvötn eruption show almost as high values as the bas_hol simulation. The time series from the Kårvatn station 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 $PM_{2.5}$ concentrations. The $PM_{2.5}$ 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^2~~ m^{-2} 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_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, Gislason et al., 2015). Model simulations with the EMEP~~/~~ MSC-W model with emissions from Holuhraun over the period from September to November 2014 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_2. Two sensitivity model simulations with different Holuhraun emission height are compared to the satellite data together with the ~~basic~~best guess base simulation. After the kernel is applied, the results are more comparable to the satellite data. ~~The results also show that it is difficult to conclude if~~Constraining the ~~discrepancies are due to~~SO_2 column burden by the ~~concentrations or~~satellite data, while using the ~~vertical placement.~~ kernel along with the three simulated height distributions of SO_2, we estimate that the median of the daily burdens may have been between 13 and 40 kt in the North Atlantic area under investigation.

The model simulations are also 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. There is a better timing in the two model simulations where the emissions are injected lowest into the atmosphere, than for the sensitivity run with the highest emission height. Due to ~~the~~a special transport pattern of SO_2 during the first event, observed concentrations are higher here than during the later events, and the difference between models and observations is largest. $PM_{2.5}$ concentration during this first 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~~To make a better ~~estimate~~assessment 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~~stations studied and other stations ~~showed~~to be found in the ~~appenidx~~supplementary material.

Studying the changes in pollution levels over Europe, increased SO_X wet deposition ~~showed~~is most remarkable. In the ~~highest increase in the model. For the basic~~three month base simulation there is 32 % more sulphate wet deposition found than in the model simulation with no Holuhraun emission, accounted for 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%, shows levels equal to the most polluted regions in Europe ~~Compared with~~ Seen against the long-term record of observed levels of deposition since 1980 at the Tustervatn station in central Norway, the 2014 ~~model~~deposition values stand out and are ~~earlier~~ only ~~reached in the observations~~exceeded during the Grimsvötn eruption in 2011. ~~Higher~~We also find that high SO_X wet deposition values measured at the Kårvatn station in 2014 on the coast of western Norway are very likely 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_2 concentrations over Europe between the no_hol and model simulations with Holuhraun emission ~~are~~is around 13 percent ~~over the same 30 countries~~ and increases ~~occurs~~occur 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~~simulated volcanic contribution is possibly too small. For $PM_{2.5}$ concentration, the volcanic increase is six percent, and the model shows better agreement with station observations. The biggest difference in percent increase 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 an increase in number of days with over 25 ~~$\mu g/m^2$~~$\mu g m^{-2}$ $PM_{2.5}$ concentrations are those already polluted. Even with high emissions from the volcanic fissure at Holuhraun, the increase in pollution levels over Europe ~~is~~was relatively small, with only transient episodes associated with high increases in SO_2 concentration.

Acknowledgements

Most of the work done for this paper is funded by the Norwegian ash project financed by the Norwegian Ministry of Transport and Communications and AVINOR. Model and support is also appreciated ~~trough~~through the Cooperative Programme for Monitoring and Evaluation of the Long-range Transmission of Air Pollutants in Europe (No: ECE/ENV/2001/003). The observations are made available trough the EEA UTD database (http://fme.discomap.eea.europa.eu/fmedatastreaming/AirQuality/AirQualityUTDExport.fmw) and the MACC project (MACC III project number 633080) obtained with the much appreciated help of Álvaro Valdebenito. Wenche Aas and Paul Eckhardt are thanked for provision of recent measurements of wet deposition measurements in Northern Norway, included in the EBAS database. This work has also received support from the Research Council of Norway (Programme for Supercomputing) trough CPU time granted at the super computers at NTNU in Trondheim.

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Table 1. Overview of model runs and the Holuhraun SO_2 emission height assumptions and flux~~.~~; given are also medians of daily mass burdens of SO_2 for September to October 2014 in the North Atlantic as described along with Fig. 2 and Fig. 3.; last column contains scaled mass burdens, assuming 7.0 kt of SO_2 burden derived from satellite data (see text in discussion).

Model ~~run~~ simulation name	~~Holuhraun layer into which SO2 was injected in the model simulation~~Emission injection layer [km]	~~Holuhraun~~ Emission flux [kg s^{-1}]	Burden original [kt]	Burden kernel weighted [kt]	Mass burden scaled [kt]
bas_hol	0 - 3 ~~km~~	750 ~~kg/s~~	22.5	7.4	21.4
low_hol	0 - 1 ~~km~~	750 ~~kg/s~~	25.4	4.4	40.3
high_hol	3 - 5 ~~km~~	750 ~~kg/s~~	15.9	8.3	13.4
no_hol		0			

Formatert: Tysk (Tyskland)

Formatert tabell

Formatert: Midtstilt, Linjeavstand: Enkel

Formatert: Midtstilt, Linjeavstand: Enkel

Formatert: Midtstilt, Linjeavstand: Enkel

Formatert: Midtstilt, Linjeavstand: Enkel

Formatert: Midtstilt, Linjeavstand: Enkel

Table 2. Emissions, depositions and concentrations for the 28 European Union member states, Norway and Switzerland for the three months (September, October, November). 2014; Emissions and depositions are totaltotalled over the three month period, concentrations are the mean over the period for the 31 countries. Numbers in brackets are the contribution from Iceland, for emission and deposition, the number represents the sum over Iceland. . For concentrationconcentrations, the number represents the average over Iceland. See simulation names and set-up in table 1.

Simulations:	no_hol	bas_hol	low_hol	high_hol	bas_hol/no_hol
Emissions SO_2	1 257	1 257	1 257	1 257	1
[kilotons]	(18)	(5 980)	(5 980)	(5 980)	(5.68)
SO_X Wet deposition	1 043	1 382	1 285	1 465	1.32
[kilotons]	(11)	(1 122)	(1 491)	(472)	(2.37)
SO_X Dry deposition	481	529	524	526	1.10
[kilotons]	(4)	(151)	(409)	(8)	(1.40)
Mean SO_2 surface conc.	1.39	1.58	1.56	1.56	1.13
[μg/m^3]	(0.59)	(38.95)	(105.91)	(1.81)	(66.17)
Mean $PM_{2.5}$ surface conc.	5.86	6.20	6.09	6.28	1.06
[μg/m^3]	(0.82)	(2.50)	(3.13)	(1.12)	(3.06)

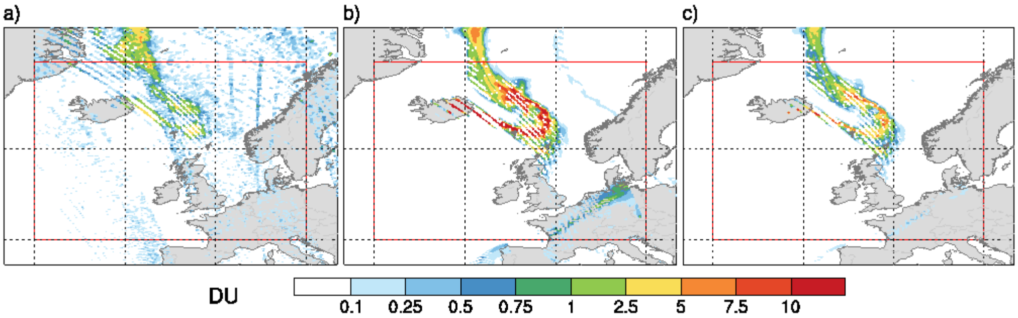

Figure 1. SO_2 column density: a) for a) the satellite swaths on 24 September, 2014; b) for corresponding, consistently co-located original model data forin the basicbase simulation from 24 September, and bas_hol; c) for these model bas_hol data with the satellite averaging kernel applied from satellite data.. The red box indicates the area where the satellite statistics of satellite and model data in fig.2 and 3 are doneacquired.

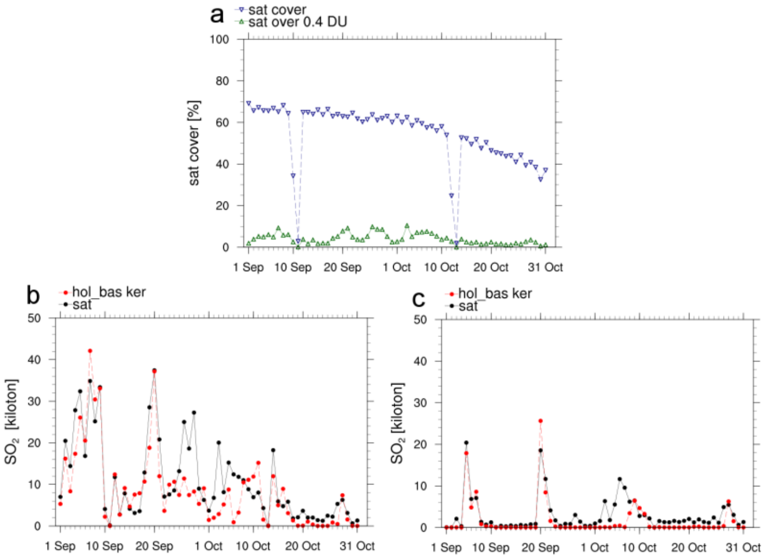

Figure 2. a) ~~Daily time~~Time series of the daily reference area covered by valid satellite ~~observed area coverage~~observations (blue triangles) in percent of the total area of the domain used for the statistics (30°W - 15°E and 45° – 70°N, see ~~fig~~Fig. 1). Green triangles show the reference area in percent ~~of the area~~ where satellite derived SO_2 VCD is above 0.4 DU~~.~~; b) ~~Daily time~~Time series of accumulated daily SO_2 mass burdens in reference area from satellite data ~~(~~("sat", black dots) and from model ~~control~~base run ~~(red dots)~~ with averaging kernel applied~~, accumulated in consistent area.~~ ("hol_bas ker", red dots; c) Shows the same as b) but over a smaller area just south of 61.15~~ degrees north~~°N.

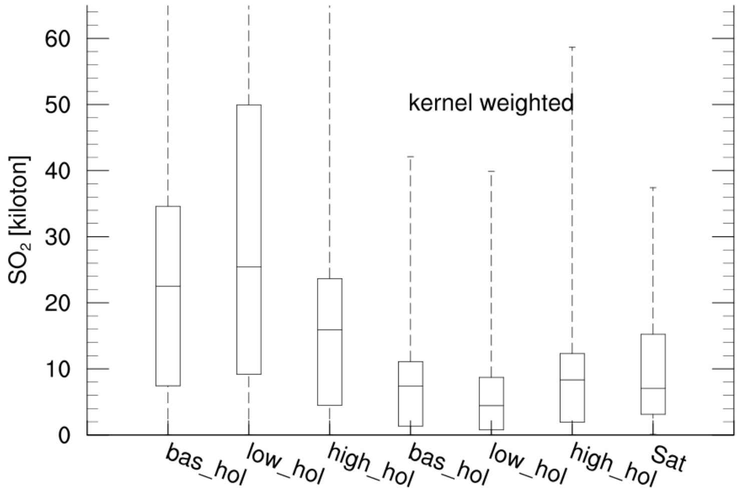

Figure 3. Distribution of daily SO_2 mass burden ~~derived from the 61 daily~~ values ~~(see fig 2) for the three model simulations, one for each of the three kernel weighted and the satellite data,~~ in the area where satellite derived SO_2 exceeds 0.4 DU (# 61; values from Fig. 2) as box and whisker plots; shown for the model simulations, original data (3 left boxes) and kernel weighted data (3 following boxes), and the satellite data. The boxes shown represent the 25th percentile, the median, and the 75th percentile values, lower whiskers the minimum value and upper whiskers the maximum value.

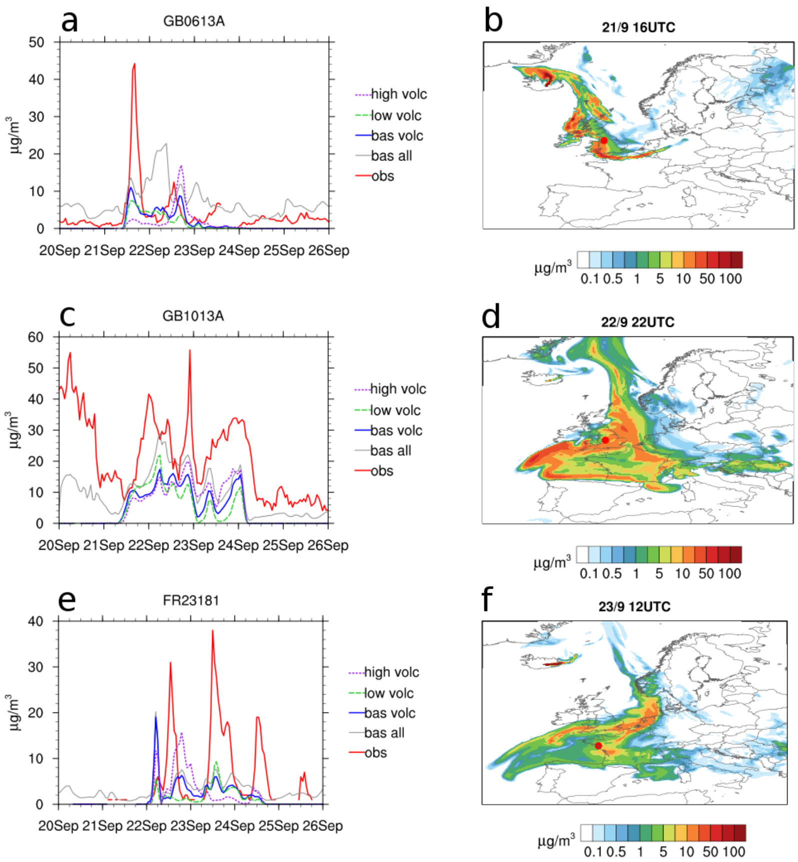

Figure 4. Left panels: Time ~~Series~~series of surface concentrations from 20 to 26 September 2014 for two stations, GB0613A~~ in~~, Manchester~~ of~~, SO_2 (top~~) and~~), $PM_{2.5}$ (~~below~~middle) and ~~SO_2~~ FR23181~~ in~~, Saint-Nazaire~~.~~, SO_2 (bottom). The red line shows the measured ~~ground~~surface concentrations, the grey line represents the modelled ~~ground~~surface concentration ~~with~~in bas_hol~~.~~ ("bas all"). By subtracting ~~the ground concentrations~~ from the modelled surface concentrations in the three model runs (bas_hol, low_hol and high_hol) the no_hol simulation values, the concentration due to volcanic eruption ~~for the bas_hol, low_hol and high_hol~~is calculated and ~~are~~ shown in the blue, green and pink line respectively. Right~~:~~ ~~Ground~~ panels: Corresponding map of simulated surface concentration due to the volcanic eruption from bas_hol, corresponding to the blue line in the ~~time series~~left panels, for the time of the maximum observed concentration. The red dot on the map marks the position of the station.

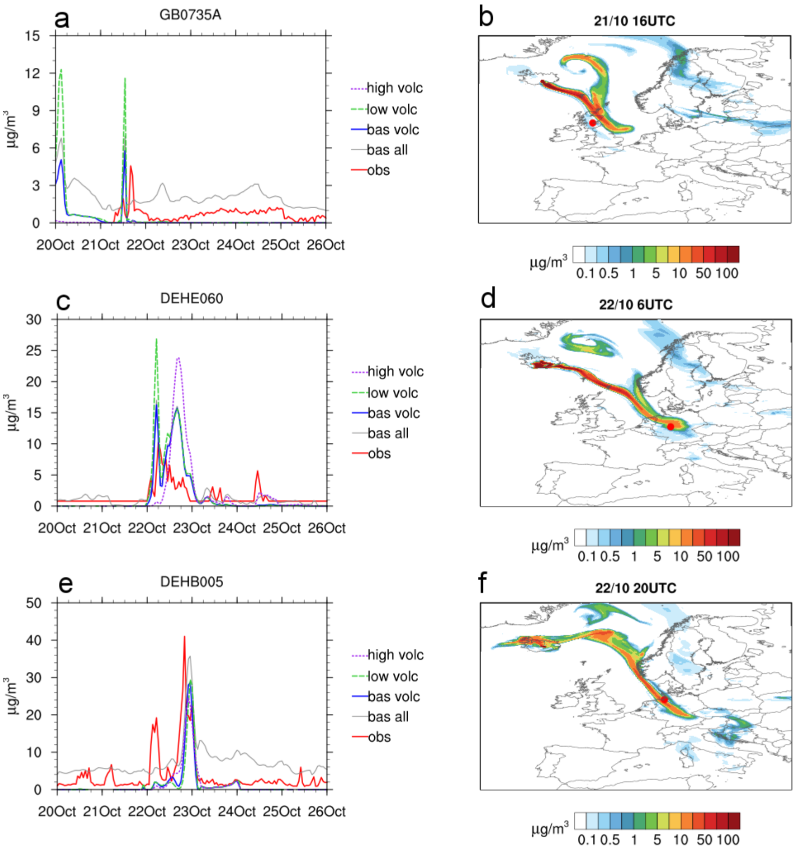

Figure 5. The same as ~~Figure 1~~Fig. 4, but from 20 to 26 October 2014 for three different stations GB0735A Grangemouth in Scotland, DEHE060 Kellerwald and DEHB005 Bremerhaven in Germany, all SO_2.

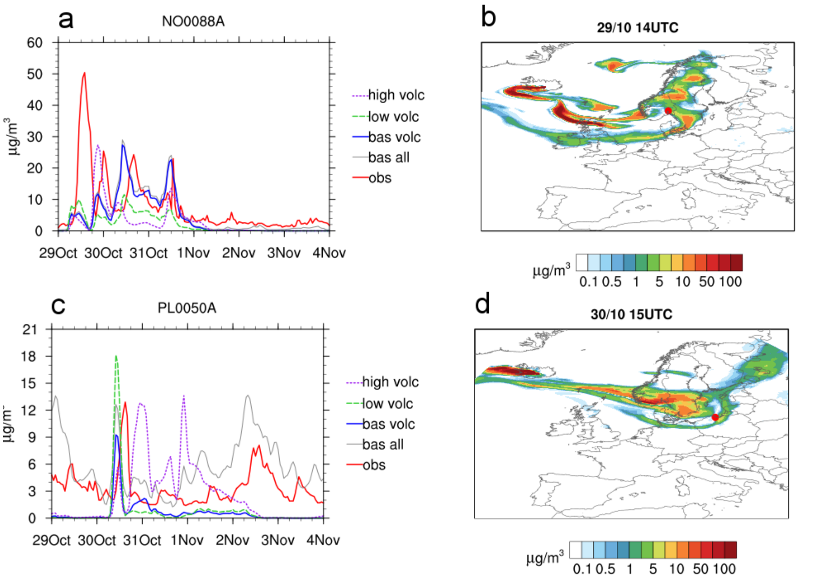

Figure 6: The same as ~~the two previous figures~~Fig. 4 but from 29 October to 4 November 2014 for NO0088A Oslo, Norway and PL0050A in Sopot Poland, both SO_2.

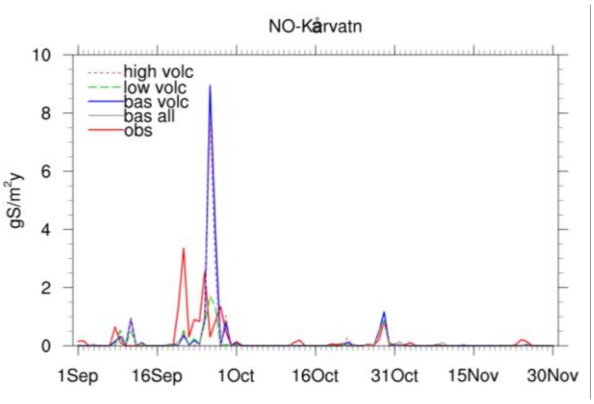

Figure 7. ~~Daily~~The same as Fig. 4, but September-November 2014 daily time series of SO_X total deposition ~~from The~~at the Kårvatn station in Norway. ~~The lines represent the same as the three plots above.~~

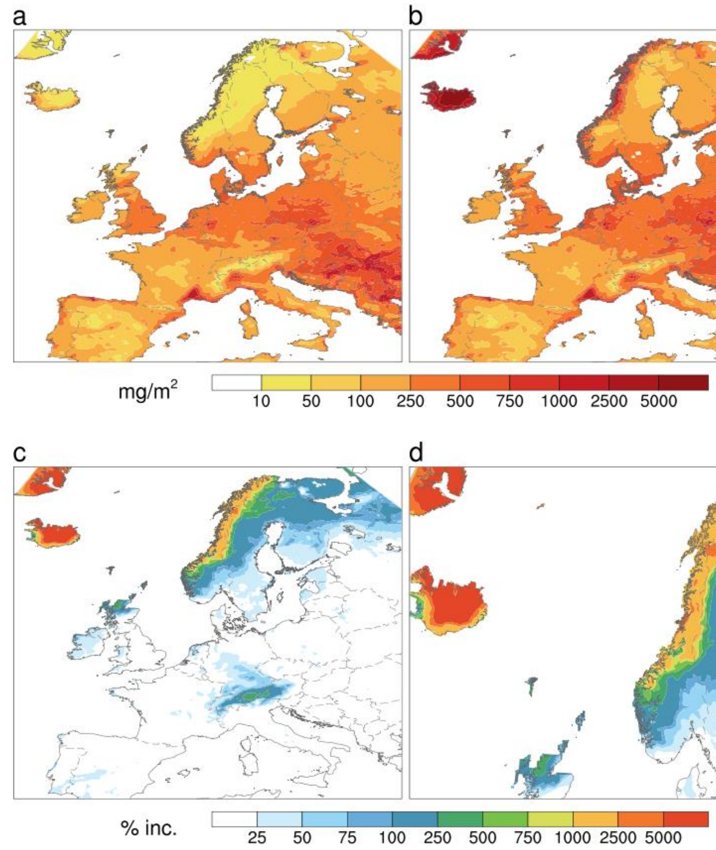

Figure 8. ~~Total~~a) Simulated total deposition of SO_X (wet and dry) over Europe from September to November 2014 for no_hol ~~(a)~~; and b) bas_hol ~~(b)~~ simulations ~~and the~~; c) percent increase in SOx deposition due to the Holuhraun emissions ~~(c).~~; d) ~~Shows~~shows the same as c) but zoomed into Norway and Northern Europe.

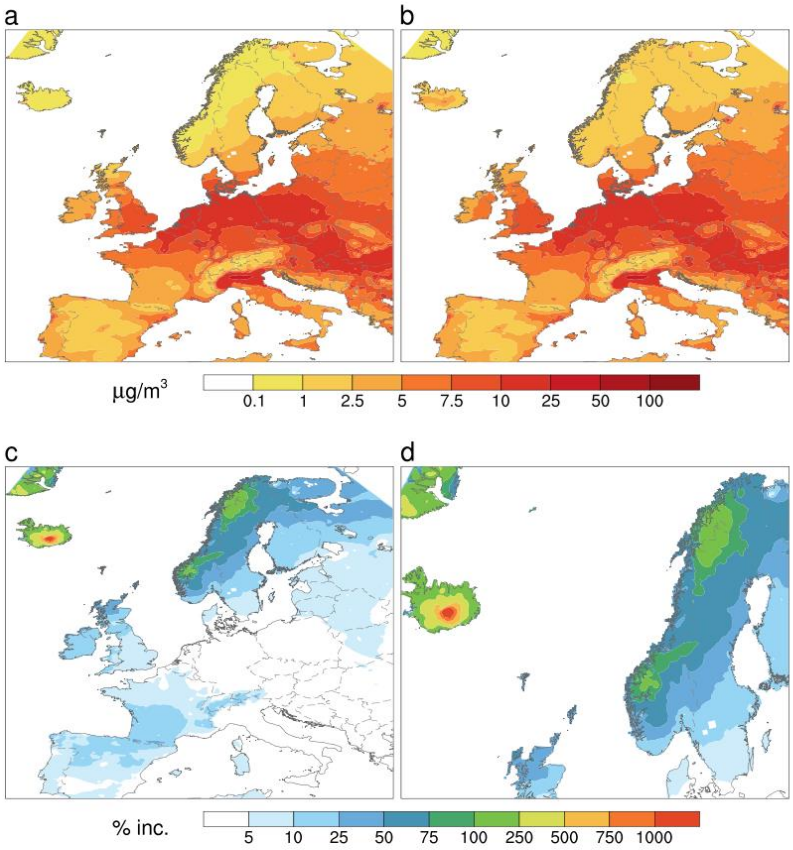

Figure 9. Show the same as ~~Figure~~Fig 8, but ~~with~~for average $PM_{2.5}$ concentration over the three months.

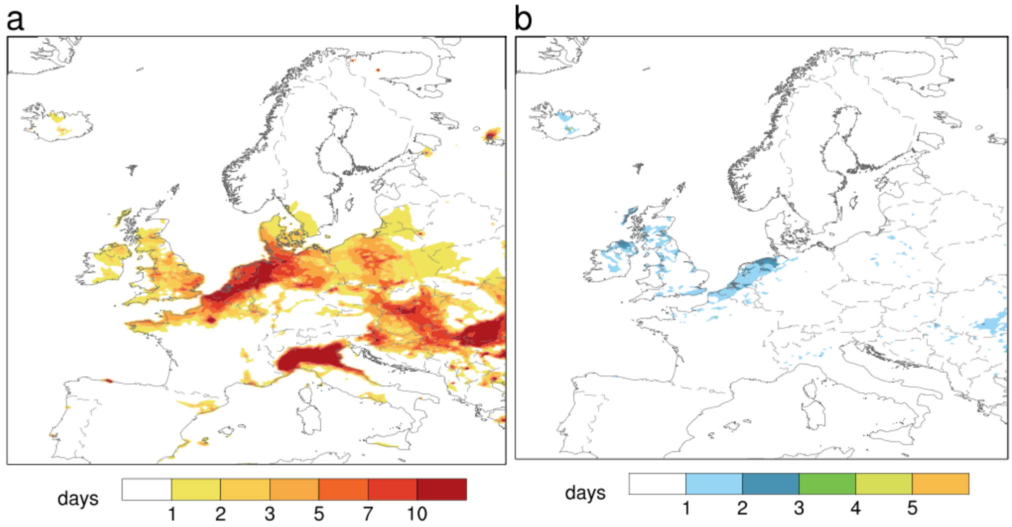

Figure 10. a) ~~Days~~Number of days with exceedances of $PM_{2.5}$ ~~over~~in the period September trough to November, 2014, for the bas_hol model simulation. b) The increase in days of $PM_{2.5}$ exceedance from no_hol to bas_hol simulation, attributable to volcanic emissions.