Dear ACP Editor and Anonymous Referees,

Please find below our answers to the 2 Anonymous Referees. In blue, the referee's comments, in black our responses.

The manuscript has been improved following the reviewers' requests, in particular note that the effect of clouds above the volcanic plume could significantly change the calculated SO_2 total emissions by 50% (from 4.4 Tg to 6.7 Tg).

At the end of the document you will find the differences between the 2 manuscripts.

Anonymous Referee #1

In the manuscript entitled "Satellite-derived sulphur dioxide (SO2) emissions from the 2014-2015 Holuhraun eruption (Iceland)", the authors derive the first timeseries of the SO2 emissions for the entire Holuhraun eruption. In a first stage, using a retrieval scheme previously developed, the authors retrieve the SO2 amount and altitude of the Holuhraun plume from IASI observations. Based on these, the authors then determine SO2 total masses every 12 hours in a large box (30_N-90_N) covering the Northern Hemisphere. They finally retrieve the SO2 fluxes for 12-hour periods using an optimal estimation scheme, considering the retrieved total masses as the measurement vector. To assess their results, the authors compare the retrieved SO2 columns, plume altitude and SO2 emissions with different type of ground-based measurements. While the SO2 emissions determined in this paper are important for different applications, a few main issues and several specific comments should be addressed before publication.

1) Page 2, lines 27-30, the authors mention that because of some geophysical conditions,

part of the SO2 plume can be missed by IASI, and thus, the derived SO2 masses should be considered as minima. This is totally true, the presence of clouds and/or low thermal contrast can hamper the detection of the SO2, and this is a complicated problem to deal with when estimating the SO2 masses. However, the authors stay very qualitative on this problem and more particularly do not mention this problem anymore in the rest of the paper (e.g. in the comparisons). It seems that the SO2 total mass derived by the authors for the entire eruption is lower than those previously estimated (Gauthier et al, 2016; Pfeffer et al., 2018; Gíslason et al., 2015; Thordarson and Hartley, 2015), but this is not discussed in terms of the underestimation of the SO2 masses and its effect on the estimated fluxes. Since the latter will be available for future comparisons and for model simulations, the authors should discuss this deeper and try to evaluate (and I realize that it is a complicated problem) how large can be the underestimation of the retrieved SO2 masses and how this underestimation affects their SO2 fluxes.

Thank you for this comment. Indeed it is not trivial to estimate the underestimation.

We have now included in the paper a way to estimate how much the SO_2 mass could be underestimated due to meteorological cloud above the SO_2 plume. This correction can be applied to other datasets that include the altitude of the plume, and is based on monthly cloud statistics from the ESA CCI project.

We also included an estimate of SO_2 plume missed due to low thermal contrast using the OMI BIRA SO_2 dataset.

Our approach is summarised as follows:

- We estimated the percent of SO₂ missing due to cloud above the plume, as a function of cloud optical depth and the altitude of the meteorological cloud above the SO₂ plume, using simulations with a standard atmosphere as done in Fig 6 of Carboni et al 2012: (https://www.atmos-chem-phys.net/12/11417/2012/acp-12-11417-2012.pdf)

- Using the ESA cloud CCI dataset of AVHRR (carried on the same platform as IASI and so having the same overpass time) L3 monthly mean statistic, we computed:

1) Monthly mean histograms of frequency of cloud optical depth (COD) at 550 nm, τ , averaged over the globe. Cloud optical depth is not present in the cloud L3 database for locations without daylight (e.g. visible channels) and most of the Icelandic plume in the winter months is without daylight, as a consequence here we are assuming the global histogram of frequency of COD is valid over the plume region.

2) Monthly mean histogram of frequency of cloud altitude, averaged on the plume region (30_N-90_N). Cloud altitude is available for all locations and during winter months.

We consider the measured mass M_{meas} to be the difference between true mass M_{corr} and the missing one M_{miss} :

$$M_{corr} - M_{miss} = M_{meas}$$
$$M_{corr} \left(1 - \frac{M_{miss}}{M_{corr}}\right) = M_{meas}$$
$$M_{corr} = M_{meas} \left(\frac{1}{1 - \frac{M_{miss}}{M_{corr}}}\right)$$

We compute the correction factor, *C*, for every month of the eruption as a function of altitude, and applied to the vertical distribution dataset.

$$M_{corr}(h) = M_{meas}(h) \cdot C(h)$$

With:

$$C(h) = \frac{1}{\left(1 - Z(h)\right)}$$

Where Z(h) is the SO₂ mass fraction 'missed' in the measurements due to cloud above the plume.

Z(h) is estimated as the product of probability of having cloud above altitude *h*, F(h), times the attenuation due to cloud, *A*,

$$Z(h) = F(h) \cdot A$$

The probability of having cloud above h has been estimate from CCI data for the region considered for the volcanic plume (latitude > 30 N) as the number of cloud retrievals above altitude h divided by number of observations.

Attenuation due to cloud (*A*) is the sum of the frequency of having a cloud with a cloud optical depth $f(\tau)$ times the attenuation due to a cloud with the same optical depth $a(\tau)$.

$$A = \sum_{\tau=0}^{n} f(\tau) a(\tau)$$

f(t) has been estimated using the monthly mean histogram of frequency of cloud optical depth, estimated over the globe.

 $a(\tau)$ has been estimated by running the SO₂ retrieval using, as IASI measurements, simulated spectra with water cloud above the plume, using the default atmosphere, and different optical depths at 550 nm. For optical depths bigger than 10 the attenuation is 1 (cloud is opaque and completely mask the SO₂ signal).

The figures below show:

- Correction factor.
- SO₂ vertical distribution obtained from IASI retrieval (was already in the paper).
- SO₂ vertical distribution corrected (for underestimation due to cloud cover).



Figure 1. Correction factor for the SO_2 masses to estimate to correct for the presence of cloud above the plume.



Figure 2. SO_2 vertical distribution in km above sea level. The colour represents the mass of SO_2 , dark-red represents values higher than the colour-bar. Every column of the plot is generated from an IASI map (one every 12 hrs). First plot show the data obtained from IASI maps, second plot is the first plot times the correction factor (to include SO2 that statistically has been missed by the IASI measurements due to cloud above the plume)

The emission fluxes have been estimated with both the original SO₂ masses (from IASI retrieval) and the masses corrected by cloud cover.

The total emission estimated with a cloud correction is 6.7 Tg (without the correction it is 4.4 Tg). These results have been added to the manuscript.

We estimate the missing SO_2 due to thermal contrast by comparison with OMI SO_2 (UV dataset) for the month of September and October 2014, as the OMI dataset doesn't fully cover the eruption time period due to the lack of solar radiance during the winter.

We visually inspected the daily maps of IASI and OMI and identified the parts of plume missing from the IASI detection (and consequently missing in the IASI retrieval).

Here is the list of all areas identified and the SO₂ estimate from OMI (BIRA-IASB)

Date	Max latitude	Min latitude	Max longitude.	Min longitude.	OMI SO ₂ for 7km height [kT]	OMI SO ₂ for 0-1 km a.g.l. [kT]
20140901	70	60	-20	-36	7.25	9.6
20140915	70	75	-10	-20	9.5	11.1
20140915	75	70	30	0	21.5	33.7
20140929	70	65	-15	-30	2	3
20140929	77	63	-20	-40	12	26

During the first 2 months we miss parted of the plume corresponding to (summing them all) 83.4 kt = 0.08 Tg (1 [Tg] = 1000 [kt])

The emission estimate (sum of fluxes * interval of time between 2 maps) from IASI for the first 2 month is 2.71 Tg, the missing mass of SO₂ (estimate with OMI 0-1 km) summed over the first 2 months is 0.08. Then the estimate of SO₂ missing due to thermal contrast is around 3%. (0.08/2.71 = 0.03).

The total mass of SO₂ missed due to thermal contrast is estimated to be few percent of the emission estimate by IASI. In particular the missing plume for the first 2 months has a total mass of 0.08 Tg of SO₂ that corresponds to 3% percent of the emission estimate by IASI.

Low SO_2 cloud also be a problem for OMI. If the OMI values are wrong by a factor 2-3, the underestimation will change to 6-9% (instead of 3%).

This estimate has been added to the text.

2) Page 3, lines 10-18, it is explained that the estimation of the SO2 masses is performed for a box going from 30_N to 90_N, considering that the SO2 detected comes from the Holuhraun eruption only. However, in this large box, other SO2 sources (China, Norilsk, volcanoes) are located and contribute to the total SO2 in this box.

While the SO2 amount emitted by these sources is probably negligible compared to the one emitted at the beginning of the eruption (masses of 0.1-0.3 Tg), I am afraid these sources could contribute to a larger percentage the days where SO2 masses lower than 0.1 Tg are estimated. For instance, the annual SO2 emissions of Norilsk are estimated to be around 2 Tg (Fioletov et al., 2016). On a daily basis, this can lead to SO2 masses around 0.001-0.01 Tg, and this can represent a large percentage of the estimated SO2 masses. This is especially the case from December, when the SO2 masses are mostly lower than 0.1 Tg. Moreover, in this period, the thermal contrast values and humidity conditions in the Norilsk area, but also in China, were shown to favour the measurement of near-surface SO2 (Boynard et al., 2014; Bauduin et al., 2014; 2016).

How did the authors take into account these extra sources? Did they remove a background of SO2 from their masses? How large do they estimate the contribution of the other sources and how does it affect the estimated SO2 masses and fluxes? This issue deserves more explanations and investigations.

We expanded the algorithm description (as also requested by referee 2) in section 2, and we hope that it is clearer now.

The IASI SO₂ algorithm is based on

1) A detection scheme that only uses the v3 band.

And for all the pixel identified by the detection

2) An iterative retrieval scheme that includes both the v1 and v3 bands.

Our detection scheme, whose theory is explained in Walker et al. (2011, 2012), is a linear retrieval with one free parameter - the column amount of SO_2 . In particular we assume the vertical distribution of SO_2 and the atmospheric vertical profiles (temperature and trace gases). We don't take into account negative thermal contrast so that regions with negative thermal contrast (such as Norilsk) often give negative values of SO_2 column amount. You can see this artefact for the month November 2013 in the following plot where negative monthly means around Norilsk are white.



Figure 3: Global IASI SO₂ linear retrieval output averaged for November 2013.

More monthly mean plots are available here (as supplementary information of the paper Taylor et al 2018):

https://agupubs.onlinelibrary.wiley.com/action/downloadSupplement?doi=10.1002%2F2017JD027109 &file=jgrd54568-sup-0002-supinfo.gif

In ours scheme we consider detection 'positive' if the output of the linear retrieval is greater than a defined positive threshold.

The full IASI dataset from 2007 to 2014 has been analysed with the same linear retrieval and results are presented in Taylor et al (2018).

Taylor, I.A., J. Preston, E. Carboni, T.A. Mather, R.G. Grainger, N. Theys, S. Hidalgo and B. McCormick Kilbride, Exploring the utility of IASI for monitoring volcanic SO₂ emissions, *Journal of Geophysical Research: Atmospheres*, **123**, 5588–5606, 2018. (doi:10.1002/2017JD027109)

The threshold for positive detection is only exceeded in the area of Norilsk when this region is affected by a volcanic plume (such as from Kasatocy, Sarakyev...). This is a limitation of our detection scheme, and improvements are under development, but for the purpose of this paper this means that Norilsk's emissions are not included as part of a volcanic plume.

The movie of the IASI SO₂ plume (present in supplementary data) shows the absence of the Norilsk contribution to the plume: there is only SO₂ detection and retrieval over the Norilsk area when a volcanic plume overpasses the area.

3) I have a few comments on the method used to estimate the SO2 fluxes. First of all, the authors should explain the advantages of the method they use compared to others ones (Theys et al., 2013). Then, I am concerned about the assumption of an averaged constant lifetime for the entire eruption. As mentioned by the authors, the lifetime of SO2 is very variable and depends on humidity, solar irradiation and altitude of the plume. Because the eruption lasted 6 months and the plume travelled very far from the source, these conditions significantly varied during the eruption and according to the location of the plume. Therefore, I am wondering why the authors have made this choice of method and why they did not consider a more sophisticated method, using a dispersion model to estimate the SO2 fluxes (Theys et al., 2013). At page 7,

line 26, the authors mention that the flux uncertainties include the possible variation of the e-folding time. This is not clear how this is done. In conclusion, the authors should justify their choice of method and provide a clear explanation of how they assess the impact of a constant lifetime on the retrieved SO2 fluxes.

We agree that the best way to estimate the fluxes will be to combine satellite measurements with a dispersion model, possibly using a scheme with a variational assimilation of the SO_2 plume height and column retrievals as we have done for Eyjafjallajokull (Vira et al 2017).

Vira, J., E. Carboni, R.G. Grainger and M. Sofiev, Variational assimilation of IASI SO₂ plume height and total column retrievals in the 2010 eruption of Eyjafjallajökull using the SILAM v5.3 chemistry transport model, *Geoscientific Model Development*, **10**, 1985–2008, 2017. (doi:10.5194/gmd-10-1985-2017)

This manuscript presents a way to derive the emission flux from a time series of total mass loading from satellite data only, and doesn't require a dispersion model. Adding this would be an enormous effort outside the scope of the paper.

We have added text to the paper to suggest further work on the use of an assimilation scheme to give the emission fluxes, the vertical distribution of emissions and associated errors.

For the first month of the eruption we compared satellite datasets (IASI and OMI) with a dispersion model (NAME) simulations (Schmidt et al 2015). Different fluxes and emission altitudes were tested to estimate the values that better match model and satellite, The fluxes found with this comparison are consistent with the values estimated here and the following figure has been now added to the manuscript.



Figure 3. Time-series of emission fluxes of SO2. To facilitate the comparison with the flux estimates from Schmidt et al. (2015), the top plot presents values (for the first month only) of: the retrieved SO2 flux time-series from IASI measured mass in black with grey error bars, the retrieved flux time-series from IASI 'corrected' mass in red with orange error bars and DOAS ground measurements from Pfeffer et al. (2018) in blue (blue bars show the errors, dotted bars show the maximum and minimum values measured that day). The bottom plot shows the 6-month time-series with the same colour coding.

The optimal estimation scheme gives a vector of parameters that we wish to estimate (the state vector) and associated errors. It is a Baysian scheme that fits the measurements and a priori knowledge of the state vector. In particular we minimize a cost function:

$$\chi^2 = [\mathbf{y} - \mathbf{F}(\mathbf{x}, \mathbf{b})]^{\mathrm{T}} \mathbf{S}_{\epsilon}^{-1} [\mathbf{y} - \mathbf{F}(\mathbf{x}, \mathbf{b})] + [\mathbf{x} - \mathbf{x}_a]^{\mathrm{T}} \mathbf{S}_a^{-1} [\mathbf{x} - \mathbf{x}_a],$$

where x is the state vector, y is the vector of measurements, F is the forward model (function of x and auxiliary data b), x_a is the *a priori value of the state vector and* S_e *and* S_a *are the measurement and a priori error covariance matrices* respectively. The output is the more probable state vector together with the a posteriori covariance matrix of the state vector S_x . The square root of the diagonal elements of S_x are the uncertainties given for the retrieved parameters. In this case the resulting uncertainties in the fluxes are a function of the errors in total mass, a priori errors and information content of the measurements.

4) Regarding among other things the previous comments, the present paper lack references, especially related to the estimation of volcanic SO2 emissions and the SO2 lifetime. The following references should be at least added:
McCoy and Hartmann, GRL 42, 10409-10414, 2015, doi:10.1002/2015GL067070;
Malavelle et al., Nature 546, 485-491, 2017, doi:10.1038/nature22974;
Lee et al., JGR 116, 2011, doi :10.1029/2010JD014758;
Theys et al., ACP 13, 5945-5968, 2013, doi :10.5194/acp-13-5945-2013;
Carn et al., J. Volc. Geoth. Res. 311, 99-134, 2016, doi:10.1016/j.jvolgeores.2016.01.002.

Thanks for these suggestions. References has been added to the manuscript.

5) Some parts of the manuscript are difficult to read and are not clear. For instance, the following paragraphs could be improved (see also specific and technical comments): Page 3, lines 3-6; Page 7, lines 24-31; Page 8, lines 1-12.

Manuscript has been expanded and rewritten in these parts.

Specific comments

-Abstract, line 5: the use of the optimal estimation to infer the SO2 emissions is not something new (Theys et al., 2013).

We don't agree. Optimal estimation (OE) has been used previously to estimate the mass loading of SO_2 (Carboni et al 2012, Clarisse et al 2008, 2012) not to estimate fluxes.

Theys et al (2013) use optimal estimation (in section 3.4 'inversion modelling method') to fit a dispersion model to observations.

Here we use optimal estimation to fit the measured time series of total mass loading with a forward model that reproduces the time series of total mass loading (a function of emission flux and SO_2 e-folding time)

This forward model (eq 4 in the manuscript) is the inverse of equation 6 in Theys et al 2013 (section 'Delta-M method'). Both equations derive from the solution of the same differential equation:

 $dM(t) / dt = F(t) - k \cdot M(t).$

In Theys at all (2013) the fluxes are obtained assuming the e-folding time, here the fluxes and the averaged e-folding time and their uncertainties, are estimated simultaneously based on OE.

-Abstract, line 6: "The algorithm is used to estimate SO2 fluxes of up to 200 kt: ::". This sentence sounds weird to me. I understand that the algorithm cannot estimate fluxes larger than 200 kt.

'The algorithm is used to estimate SO₂ fluxes of up to 200 kt per day and a minimum total SO₂ erupted mass of 4.4 ± 0.8 Tg'

Has been replaced with

'For the six months eruption studied, the SO_2 flux was observed to be up to 200 kt per day and the minimum total SO_2 erupted mass was 4.4 ± 0.8 Tg'

-Abstract, line 8: you say that you compared your results to model simulations. Do you refer to the comparison with the work of Schmidt et al. (2015)? You should rephrase the sentence because, the way it is written, the reader understands that you do model simulations.

'Where comparisons are possible, these results broadly agree with ground-based near-source measurements, independent remote-sensing data and model simulations of the eruption.' Changed in:

'Where comparisons are possible, these results broadly agree with ground-based near-source measurements, independent remote-sensing data and values obtained from model simulations (Schmidt et al 2015).'

-Page 2, line 12: can you specify what coverage? Is it temporal, spatial or both?

Both.

'Retrievals of SO₂ amount from Metop-A satellite are binned and averaged for successive 12 hour periods to give coverage for the entire period of the eruption.'

Changed in:

'Retrievals of SO₂ amount from Metop-A satellite are binned and averaged for successive 12 hour periods to give global coverage twice a day for the entire period of the eruption.'

-Page 2, line 22: did you use IASI data of level 1b (not apodized)? Or 1c (obtained after apodization)?

1c Apodized, this has been added to the text.

-Page 2, line 24: Can you briefly remind what is a positive result in the SO2 detection scheme?

An output values (of the linear retrieval) higher then 0.45 effective DU. Description of the scheme has been expanded including this.

-Page 2, line 28: I would specify that you miss part of low-altitude SO2 plumes in case of low thermal contrast. On the same line, you say that IASI can miss part of the SO2 plume in case of negative thermal contrast conditions. Why? It has been shown by Bauduin et al. (2014, 2016) and Boynard et al. (2014) that negative thermal contrasts are favourable conditions to measure SO2 close to the surface.

The referee is right, it is not IASI, but the IASI detection scheme used in this manuscript, that can miss part of the plume.

Session 2 has been expanded including this.

-Page 3, line 2: Why cannot you use the _3 band to measure SO2 down to the surface? Is the band saturated? Or is it because of large humidity close to the surface? Can you please explain? If none of these two reasons is true, you should be able to measure SO2 close to the surface using the _3 band (Bauduin et al., 2014; 2016).

Sentence in the manuscript:

'All the channels in the ranges 1000-1200 and 1300-1410 cm-1 (the 7.3 and 8.7 μ m SO₂ bands) are simultaneously used in the iterative optimal estimation retrieval scheme to obtain the SO₂ amount, the altitude of the plume and the surface temperature. The SO₂ band around 8.7 μ m (1000 to 1200 cm-1) is within an atmospheric window. This allows the radiation from the surface to reach the satellite from deep within the atmosphere enabling the retrieval of SO₂ amount down to the surface.'

You can use only the v3 band but, for standard atmosphere conditions, your measurements will not be affected by SO_2 close to the surface, due to strong water vapour absorption. The v3 band will only allow SO_2 retrieval close to the surface in dry condition as stated in Bauduin et al. (2014; 2016). Using v1 band together with the v3 band increases the information content of the measurements so that SO_2 is measured in all water vapour conditions.

-Page 3, line 3: How did you build the error covariance matrix? Is it a global one or did you build one more specific for the eruption? This should be explained.

We used the global covariance matrix, this has been added to section 2.

-Page 3, line 6: Can you specify what is your quality control?

Quality control (now added to section 2) is:

Cost function < 10, retrieved pressure between 0 and 1100 mb, positive SO_2 column amount, plus convergence of the iteration algorithm.

-Page 3, line 7: You say that the SO2 retrieval is not affected by an underlying cloud. However, this cloud has to be taken in the retrieval, at least in the radiative transfer. How did you take into account the underlying clouds? How did you detect underlying clouds?

We don't detect cloud, the variability of the spectra due to cloud presence is included in the measurement error covariance matrix.

We build up the measurement error covariance matrix with the differences between our forward model (radiative transfer with no cloud, driven by ECMWF profiles) and the IASI measurements. In

this way the measurement error covariance matrix include the variability of all the parameters that are not retrieved and not well represent by the forward model. The biggest contribution to this covariance matrix is the presence of cloud.

More detail on the retrieval scheme in Carboni et al (2012).

Moreover comparisons with CALIPSO measurements (Carboni et al. 2016) for cases strongly affected by underling cloud show consistency between the IASI retrieved altitude and CALIPSO backscattering profiles.

-Page 3, line 11: it is not clear to me what you combined. I suppose you created AM and PM maps each day?

Yes.

In the manuscript:

'The retrieval results from the Metop-A orbits during the period from September 2014 to February 2015 and from 30° N to 10.90° N are combined to produce two maps per day of retrieved SO₂ amount and altitude.'

-Page 3, lines 15-18: you say that you cannot make the distinction between the Holuhraun plume and the other SO2 sources, but then you make the distinction for the 21st and the 31st December. How did you do this distinction? How did you take this into account in the evaluation of the SO2 masses and fluxes? (major comment 2)

'Satellite observations at the pixel level do not provide sufficient information to distinguish between SO_2 from Holuhraun and SO_2 from other sources.'

The following comments in the manuscript (below) describing other sources come from the visual inspection of the sequence of daily maps (show as a movie in the supplemental material).

'For example, the elevated SO_2 near Beijing on 21st December 2014 appears to be from an anthropogenic source but the elevated SO_2 in the same area on 31st December 2014 is from the Holuhraun eruption. '

As is possible to see in the supplement material movie, in the maps before 21 December there is no presence of volcanic plume moving toward China, while in the maps before 31 December we can follow the evolution of the Icelandic plume over Asia and reaching Beijing.

-Page 3, line 24: the reference Boichu et al. (2015) should be added.

Done

-Page 3, line 30: can you specify how you calculate the errors on the total masses?

'The SO₂ mass present in the atmosphere for each IASI overpass was found by regridding the observations of column amount and plume altitude into a 0.125° latitude/longitude boxes following Carboni et al. (2016). The SO₂ mass time-series is obtained by summing the mass values of the regularly gridded map for each 12 hour period. The time-series of SO₂ mass, together with the errors, are presented in the top plot of Figure 2'

Changed into

'The SO_2 mass present in the atmosphere for each IASI overpass was found by regridding the observations of column amount and plume altitude into a 0.125° latitude/longitude boxes following Carboni et al. (2016). The SO_2 mass time-series is obtained by summing the mass values of the regularly gridded map for each 12 hour period. The same procedure has been used for the errors, this means that the sums of errors of grid boxes are considered as errors on the total masses (this could be an error overestimation but we cannot consider the usual errors in quadrature due to the possible presence of systematic error, e.g. the errors are not independent). The time-series of SO_2 mass, together with the errors, are presented in the top plot of Figure 2'

-Page 6, lines 10-11: why did you choose these a priori values? Did you rely on the previous literature?

'The a priori values used were 0.2 ± 0.2 Tg/day for flux and 2 ± 2 day for the e-folding time.'

For the flux we choose 0.2+- 0.2 Tg a day for a priori flux because this allows the retrieval to move easily (if there is enough information in the measurements) between 0 and 0.4 Tg/day and 0.4 is greater than the maximum value of total mass. Only a few values of total mass in September exceed 0.2 Tg and in particular none of them show a total mass greater the 0.3.

We think that the a priori e-folding time between 0 and 4 will cover the real lifetime of SO_2 for low tropospheric plumes. We experimented with longer e-folding time but the resulting fitting is worse. Assuming shorter e-folding time results as good a fit as the one presented in the manuscript, this is why we have written this at line 28 page 7: 'Also note that any e-folding time shorter than the retrieved one can fit the measurements and give higher fluxes.'

-Page 7, line 7: you did not explain what Se you considered. Can you specify it?

Not sure I understand the comment here.

Page 7 line 7 states:

'where λ (with $\lambda = 1/k$) is the average e-folding time. Equation 4 is the forward model F(x).' We define Se in pag 6, line 9-10: 'Se and Sa are diagonal matrixes with the variances (square of errors) of y and xa respectively as their diagonal elements.'

-Page 7, line 13: The averaged fluxes reported for December, January and February are very low. The impact of other sources is in this case non negligible (major comment 2). Did you take this into account?

We did consider all the SO₂ as Icelandic source. See answer to major comment.

-Page 7, lines 14-15: You did not give a tentative explanation for the fact that 1) the monthly averages of IASI fluxes are lower than those calculated from ground-based observations, and 2) the maximum values are larger for IASI than for ground-based observations. Is this because of the underestimation of the masses? Or variations in the lifetime? Or the inclusion of other sources? I think you can extend the discussion.

Here is the paragraph from the manuscript: 'The estimates for December, January and February show decreasing flux with monthly averages of 0.016, 0.006 and 0.005 Tg/day respectively (0.026, 0.028, 0.016). The monthly averages are lower than those measured by the ground-based measurements while the maximum daily averages for each month are generally higher.'

We added this ti the text:

'The UV ground-based measurements for the dark months of December, January and February are sparse, with only 10 measurements over these 3 months. There was only one day with measurements at the beginning of December, and then 6 days with measurements in the second half of January and three days with measurements in February. The extrapolated flux from the ground-based measurements through December to the first half of January is consistent with the error bars from the IASI estimates. The differences in monthly means between the ground-based measurements and the IASI flux estimates in the sunny months are explainable by low values with large error.'

-Page 7, line 16: You compare the SO2 fluxes you determined with the modelled fluxes of Schmidt et al. (2015). Why didn't you also compare your emissions with the fluxes they determined from OMI and IASI observations?

In Schmidt et al 2015 we did not estimate fluxes from IASI and OMI, we estimated fluxes comparing maps of SO_2 column amount from IASI, OMI and NAME. The NAME simulation that best matched the satellite was used to estimate the flux range reported in Schmidt et al (2015). We added the Schmidt et al 2015 fluxes estimate in fig 2.

-Page 7: I would add the errors of the fluxes in the text.

I'm not sure what this comment refer to but I guess the referee refer to these lines 15-17 at pag 7: 'The fluxes calculated for September 2014 are consistent with Schmidt et al. (2015) (e.g. up to 0.120 Tg/d during early September, 0.02-0.6 Tg/day between the 6th and 22nd of September, 0.06-0.120 Tg/day until the end of September).'

We now added a 'zoomed' plot with fluxes for September (only) including IASI estimate of fluxes, IASI error-bars and fluxes estimate from Schmidt et al. (2015) that show the consistency of IASI fluxes with Schmidt et al. (2015).

-Page 7, line 18: you calculate the total mass of the eruptions from the SO2 emissions you derived. These emissions are strongly affected by the fact that you use a constant averaged SO2 lifetime. Wasn't it more accurate to use the daily masses you estimated (even though they are underestimated)?

Summing all the daily masses, and considering this sum as the total emission, would assume that the plume in one retrieval is completely gone in the following retrieval after 12 hours. This is not true as we can follow the plume evolving in time and reaching different parts of the northern hemisphere and we can visually track the same part of plume through consecutive retrievals for multiple days. We need an estimate of flux that takes into account an e-folding time. In this manuscript we chose to take into account one e-folding time with a big a priori error that include all the e-folding time variability.

-Page 7, lines 32-33: the "spikes" you see in the SO2 fluxes, are they real? Or do they come from the forward model you used? The Delta-M method is known to produce spikes in time-dependent fluxes (Theys et al., 2013).

(Theys et al., 2013) state that (in the Delta-M section):

'As these series (referring to total mass series) do contain some uncertainty, the resulting flux curves often display spikes that are likely not related to real source variation.'

Delta M spikes are mostly coming from incomplete coverage of the plume, eg due to orbital position. In the case of the Holuhraun eruption we have complete coverage of the plume using IASI data.

Nevertheless it true that the fluxes estimated in this manuscript show results with some spikes but the fluxes obtained here have to be considered together with their error estimates. Here we consider the time series of total masses and associate errors in a comprehensive optimal estimation scheme.

Note that the errors in the resulting fluxes are often of the order of 100%. In figure 2 of the manuscript the grey colour band represent the flux errors and this grey band rarely is detached by the horizontal line of 'zero' flux line, even in correspondence of the spikes. It is plausible that spikes in the SO_2 fluxes are genuine.

-Page 8, lines 8-10: How do you explain that Gauthier et al. (2016) estimated lower SO2 daily masses but a higher total SO2 mass? Is this related to their choice of lifetime? This comparison should be discussed deeper.

Although is not 100% clear, from the Gauthier et al. (2016) paper, how they estimate the total emitted mass (from sparse fluxes only in correspondence of 'positive gradient' between 2 successive SEVIRI masses), there are 2 possible explanations for disagreement.

One explanation could indeed be the assumed SO_2 lifetime, but Gauthier et al. (2016) estimate that this could only affect their masses by 1.3–5.2% percent (Gauthier et al 2016, section 3.1).

The other explanation is mainly related with the fact that they use a less sensitive instrument (SEVIRI) and an algorithm that, in case of no valid measurements, interpolates between valid flux estimates.

This paragraph, that was in the manuscript:

'Had a less sensitive instrument been used that only produced 'valid' measurements in correspondence with higher flux values (e.g.> 0.05Tg/day) and had considered these fluxes as representative of the period without valid measurements (i.e. period between two 'valid'

measurements), this would result in very different (and higher/overestimated) estimated values of total SO_2 emitted. An example of this is Gauthier et al. (2016) where they used TIR data from SEVIRI, on board the geostationary satellite Meteosat Second Generation (MSG), to retrieve an SO_2 mass timeseries from 1 September 2014 to 25 November 2014. Their retrieved mass values are lower compared to the IASI values here (due to the smaller geographic area considered and possibly due to a smaller sensitivity or detection threshold of SEVIRI), nevertheless they estimate a total SO_2 emitted mass of 8.9±0.3 Tg for the period September 2014 to November 2014, which is a factor of two higher than calculated here.'

Have been substitute with this one:

'Gauthier et al. (2016) used TIR data from SEVIRI, on-board the geostationary satellite Meteosat Second Generation (MSG), to retrieve an SO₂ mass time-series from 1 September 2014 to 25 November 2014. Their daily retrieved mass values are lower compared to the IASI values reported here due to the smaller geographic area considered and possibly due to the lower sensitivity of SEVIRI. Nevertheless they estimate a total SO₂ emitted mass of 8.9±0.3 Tg for the period September 2014 to November 2014, which is a factor of two higher than IASI. Our understanding is that the Gauthier et al. (2016) scheme produced 'valid' measurements when the SO₂ loading increased with respect to the previous measurement. This leaves data gaps when the SO₂ measured remains constant or decreases. The resulting data gaps were filled by linearly interpolating between two 'valid' measurements. This has a tendency to bias flux estimates in favour of increasing SO₂ loading. The dataset of Gauthier et al. (2016) contains several days with no valid fluxes estimates and for these data gaps the interpolation of valid measurements into data gap could account for their discrepancies with our dataset.'

-Page 8, lines 11-12: Following the previous comment, I think the comparison could be extended. You did not compare the total mass you estimated with those reported by Schmidt et al. (2015) (2 Tg for September 2014) and by Gíslason et al. (2015) (11 Tg). Moreover, you should compare the total masses calculated for a same period.

The comparison should mention the difference in sensitivity, in lifetime,: : :

The total masses reported by Schmidt et al. (2015) are relative to a smaller area that the one considered here (summed from a lat-lon box of 60°W-40°E and 75°N-45°N) and in particular the IASI total masses are coming from the same IASI dataset. We did add the comparison of the fluxes estimate from Schmidt et al. (2015) in a new figure of the paper (reported as fig 3 in this document)

-Page 8, lines 17-18: As already mentioned above, negative thermal contrasts have been shown to increase the sensitivity to near-surface SO2 (Bauduin et al, 2014; 2016; Boynard et al., 2014).

See discussion above and rewritten section 2.

-Page 8, line 34: Can you explain why you compare ground-based measurements with the average of all IASI pixels located within 200 km? Why not taking the closest IASI pixel?

Due to variability of the volcanic plume we can have strong variation in space (in satellite maps) that are seen as variation in time (in ground measurement when the plume with different loading overpass the ground location at different time). It is common procedure to average satellite data over some distance from the ground location and to compare this with ground measurements taken during a time period. It is essentially assumed that variability in time is related to variability in space.

-Page 9, line 9: You say that for some days, SO2 is detected by only one instrument because the limit of detection of the other instrument is not exceeded. Is it really true? Did you check that the fact that you consider a circle of 200 km radius around the ground-based station for calculating the IASI average does not play a role (i.e. IASI can detect SO2 in a part of the circle far away from the Brewer)?

Yes we did look into everyone of the 200 km circle IASI data, and this is why we describe this in the manuscript:

'All the 'plume' episodes (with SO₂ amount larger than 2 DU) are consistent between the two datasets with the exception of 15th and 19th September where the plume only passes over the northern part of the 200 km circle in the IASI data and does not pass over the ground measurement station.'

In attach here a slide with fig 4 of the manuscript together with some of the IASI map (of column amount [DU] in colour, Day from 1st Sept of the IASI measurements in the titles) that visualized the IASI plume pixel within 200km. This shows the case of 15 and 19Th September IASI overpass where the plume detect by IASI doesn't overpass the Brewer location.



-Page 10, line 2: Can you define what is IMO (indicated in Figures 2 and 3)?

IMO = Icelandic Met Office. IMO dataset were referring to the ground-based measurements described in (Pfeffer et al., 2018). We now removed 'IMO' and replaced with DOAS for the figure with fluxes and 'observation' for the altitudes.

-Page 10: Could you specify what are the errors on altitude and SO2 fluxes calculated from ground-based measurements? How were they calculated?

Ground base measurements are described in (Pfeffer et al., 2018).

-Page 10, line 15: you say that IASI values can be underestimated. Is it because of low thermal contrast? Did you check the values?

Yes. The plume close to the surface could be underestimate due to thermal contrast.

-Page 10, lines 16-18: Since you linearly interpolated the fluxes, you overestimate the total mass when integrating below the red line (especially if the variations of IASI are real). Calculating the total mass on periods where you do not need to interpolate might improve the comparison (when the mass is compared with the one of IASI calculated for the same period).

The reviews are right but, and the interpolation (or extrapolation of fluxes where there is no measured data) is a key factor that could produce discrepancies. The problem is that we mainly need to interpolate at any time, at the best the ground measurement are performed during daylight and IASI measurements cover all the 24 h. To show the difference in time sample see fig 3 of this document that show a plot of different fluxe estimates for September only (the month with more ground measurements). This plot has been added to the paper (to include the comparison with Schmidt fluxes estimate)

-Page 10, conclusions: I find the conclusions a bit too short. I would say a word about some of your limitations (underestimation of the masses), about the comparison of the total mass,: : :

We added this paragraph into the conclusion:

'By comparison with OMI dataset we estimate that the SO_2 masses missed due to low thermal contrast is of the order of few percent (3%) of the total emission.

We did estimate the SO₂ mass missed, due to cloud above the SO₂ plume that is masking the signal, using AVHRR cloud CCI dataset monthly mean statistic. Results show a correction factor increasing with decreasing altitude, from one (no underestimation of SO₂ masses) up to a factor two (we measure 50% of the 'true' mass) for plume between 0-1 km. Appling this correction result in a total mass, emitted during the 6 month of eruption, of 6.7 ± 0.4 Tg and little change in the average e-folding time (2.5 ± 0.7). The IASI fluxes data reported here are representative of ~12 hours and with no datagaps but when comparing with different source of emission estimate the interpolation (or extrapolation of fluxes) where there is no measured data, is one of the key point that could produce discrepancies.'

The following comments have been taken into account in the new version of the manuscript: Technical comments

-Abstract, line 4: remove the comma after "data".

-Abstract, lines 9-11: The last sentence is very long and hard to read. You should rephrase it.

-Page 2, lines 2-5: I think you should rephrase the end of the paragraph, it does not read well.

-Page 2, line 11: I would replace "the first time series of the Holuhraun SO2 plume" by

"the first time series of the Holuhraun SO2 emissions".

-Page 2, lines 16-18: I would rephrase the last sentence, it is a little bit too long.

-Page 2, line 21: add "a" before "sampling" and "almost" before "global".

- -Page 2, line 26: "are estimate" ! "are estimated".
- -Page 3, line 30: Replace "The SO2 mass is highest" by "The largest SO2 mass is found".
- -Page 6, line 6: "y" should be bold.
- -Page 6, line 9: matrixes ! matrices.
- -Page 7, line 28: "then" ! "than".
- -Page 8, line 6: Gauthier et al. (2016) is not included in the references at the end of the manuscript.
- -Page 8, lines 21-24: I found this sentence very long and difficult to read. I would

rephrase it.

-Page 10, line 1: "collated" ! "collected"?

-Page 10, line 6: "groud" ! "ground".

-Page 10, line 15: "underestimates" ! "underestimated".

-Figure 2, top: The blue line is difficult to distinguish from the black dots. Maybe change the colour?

Blue changed with light green. Following addition of estimate of missing SO₂ we now added both estimate of total masses and fluxes with uncorrected and corrected data. New fig 2 below.



-Figure 3: In the text, you say that some of the ground based measurements provide the altitude of the plume center-of-mass, and the others the altitude of the top of the plume. Maybe you could make the distinction between the two cases in the Figure (circle and triangle, or something else). It would be easier to see which ground-based observations provide the same information than IASI.





References

Bauduin et al., J. Geophys. Res. Atmos. 119, 4253-4263, 2014, doi:10.1002/2013JD021405; Bauduin et al., Atmos. Meas. Tech. 9, 721-740, 2016, doi :10.5194/amt-9-721-2016; Boynard et al., Geophys. Res. Lett. 41, 645-651, 2014, doi :10.1002/2013GL058333; Fioletov et al., Atmos. Chem. Phys. 16, 11497-11519, 2016, doi :10.5194/acp-16-11497-2016; Thordarson and Hartley et al. Geophys. Res. Abstracts, 17 (EGU2015-10708).

Anonymous Referee #2

This paper developed a new scheme to calculate daily SO2 fluxes and average efolding time for volcanic SO2 emissions in Iceland. In order to overcome the difficulties in latitude and time, the authors propose to use satellite-based thermal infrared spectrometers instead of UV bands to study the volcanic SO2. The results look sound and interesting. I recommend publishing the paper after addressing the comments below.

General comments:

1. Page 3, line 18. In this study all the SO2 measured from 30N to 90N between September 2014 and February 2015 is referred to as Holuhraun SO2. What is the uncertainty of this assumption?

We estimate the atmospheric loading of non-Holuhraun source as no higher than 0.01 Tg. This estimate comes from the SO_2 total mass loading during the second half of February where there is no presence of plume from Iceland. The SO_2 in these two weeks mainly comes from China and from some volcanic activities in Kamchatka.

2. This paper is based on the previous work performed by the same author. I understand the authors would like to keep the text simple and avoid repeating contents mentioned by their previous work. However, sometime the text seems to be too brief to keep all important information. For example, Page 3, line 27-28. "regridding the observations of column amount and plume altitude into a 0.125 latitude/longitude boxes following Carboni et al. (2016)." What is special of the regridding approach in Carboni et al. (2016)? I have the similar concern for Section 2.

Section 2 with the algorithm description have been expanded including more detailed explanation of the regridding

Specific comments:

1. Page 2, line 22. The exact location of the IASI data should be added.

Added.

2. Page 2, line 30. Putting a rough quantification of the uncertainty of the "minimum" here would be appreciated.

We added into the manuscript an estimate of the underestimation due to cloud cover and thermal contrast, using respectively AVHRR Cloud CCI dataset and OMI SO₂ dataset, See answer to review1.

3. Page 6, Line 10. The a priori values used were 0.2 _ 0.2 Tg/day for flux and 2 _ 2 day for the e-folding time. Is there any sources for the priori values? If not, will the fitting results be sensitive to the choices of the priori values?

As for answer to review 2:

'The a priori values used were 0.2±0.2 Tg/day for flux10 and 2±2 day for the e-folding time.' For the flux we choose 0.2+- 0.2 Tg a day for a priori flux because this correspond to allow the retrieval to move easily (if there is enough information in the measurements) between 0 and 0.4 Tg/day and 0.4 is greater than the maximum values of total masses. Only few values of total masses in September exceed 0.2 Tg and in particular none of them show total mass greater the 0.3.

We think that the a priori e-folding time variation between 0 and 4 will cover the real lifetime of SO_2 for low tropospheric plume, we experiment assuming longer e-folding time to the analysis and the resulting fitting is worse. Assuming shorter e-folding time instead result in good fit as well as the one presented in the manuscript, this is why we have written this at line 28 pag 7: 'Also note that any efolding time shorter than the retrieved one can fit the measurements and give higher fluxes.'

4. Figure 2. The color of blue is difficult to see.

Changed with light green. See new plot in answer to referee 1.

Satellite-derived sulphur dioxide (SO₂) emissions from the 2014-2015 Holuhraun eruption (Iceland)

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Abstract. The six-month-long 2014-2015 Holuhraun eruption was the largest in Iceland for 200 years, emitting huge quantities of sulphur dioxide (SO₂) into the troposphere, at times overwhelming European anthropogenic emissions. Weather, terrain and latitude , made continuous ground-based or UV satellite sensor measurements challenging. Infrared Atmospheric Sounding Interferometer (IASI) data , is are used to derive the first time-series of daily SO₂ mass and vertical distribution over the entire

- 5 eruption period. A new optimal estimation scheme is used to calculate daily SO_2 fluxes and average e-folding time every twelve hours. The algorithm is used to estimate For the six months studied, the SO_2 fluxes of flux was observed to be up to 200 kt per day and a the minimum total SO_2 erupted mass of was 4.4 ± 0.8 Tg. The average SO_2 e-folding time was 2.4 ± 0.6 days. Where comparisons are possible, these results broadly agree with ground-based near-source measurements, independent remote-sensing data and model simulations of the eruptionvalues obtained from model simulations from a previous paper.
- 10 The results highlight the importance of high-resolution time-series data to accurately estimate volcanic SO_2 emissions. The SO_2 mass missed due to thermal contrast is estimated to be of the order of 3% of the total emission when compared to measurements by the Ozone Monitoring Instrument. A statistical correction for cloud based on the AVHRR cloud-CCI dataset suggested that the SO_2 mass missed due to cloud cover could be significant, up to a factor of two for the plume within the first kilometre from the vent. Applying this correction results in a total erupted mass of 6.7 ± 0.4 Tg and little change in average
- 15 <u>e-folding time. The</u> dataset derived can be used for comparisons to other <u>ground</u> and satellite-based measurements, and to petrological estimates of the SO_2 fluxas well as. It could also be used to initialize climate <u>models</u>model simulations, helping to better quantify the environmental and climatic impacts of <u>future Icelandic fissure eruptions and simulations of past</u> large-scale flood lava eruptions.

1 Introduction

Sulphur dioxide (SO_2) is one of the most important magmatic volatiles for volcanic geochemical analysis and hazard assessments due to its low ambient concentrations, abundance in volcanic plumes, and spectroscopic features. Tropospheric volcanic SO_2 and its conversion products can affect the environment, human health, air quality and the radiative balance of the Earth

- 5 (Gíslason et al., 2015; Schmidt et al., 2012; Gettelman et al., 2015; Ilyinskaya et al., 2017; Boichu et al., 2016) (Schmidt et al., 2015; Gísl Measurements of SO₂ from volcanic eruptions are vital both to understand the underlying volcanic processes and also the wider scale environmental impacts of volcanism. The Icelandic Holuhraun eruption lasted from 31 August 2014 to 28 February 2015 and produced the largest lava volume in Iceland for more than 200 years (Gíslason et al., 2015). During September 2014, Holuhraun's average daily SO₂ emission exceeded daily SO₂ emissions from all anthropogenic sources in Europe by a factor
- 10 of three (Schmidt et al., 2015 and references therein). The weather conditions and terrain made regular ground-based plume measurements extremely challenging during the winter months (Pfeffer et al., 2018). The high latitude of the eruption meant there was insufficient sunlight to reliably detect the volcanic plume using UV satellite sensors beyond the end of the October 2014 and ground based 2014. Due to insufficient sunlight, ground-based UV instruments did not measure SO₂ during the darkest seven weeks of winter. Under these circumstances satellite-based thermal infrared spectrometers are an optimal source
- 15 of high temporal resolution SO_2 amount and altitude.

The Infrared Atmospheric Sounding Interferometers (IASI) on-board the Metop satellite platforms provide several observations of Holuhraun each day. The plume altitude and SO₂ column amount are retrieved from the measured top-of-atmosphere spectral radiance (Carboni et al., 2012). For the first month of the Holuhraun eruption, previous studies have shown good agreement between IASI measurements and those from the Ozone Monitoring Instrument (OMI), ground-based and balloon-borne

- 20 measurements, and atmospheric dispersion model simulations (Schmidt et al., 2015; Vignelles et al., 2016). In this work IASI measurements are used to produce the first time series time-series of the Holuhraun SO₂ plume. Retrievals of SO₂ amount from the Metop-A satellite are binned and averaged for successive 12 hour periods to give coverage global coverage twice a day for the entire period of the eruption. The time-series of the SO₂ mass present in the atmosphere is used to calculate SO₂ fluxes and an average SO₂ e-folding time, under the assumption that the flux is constant over a twelve hour-12-hour period.
- 25 The results are compared with ground-based Brewer measurements of the SO_2 column amount and with measurements of near-source plume altitudes and fluxes from the Icelandic Meteorological-Office (IMO). The dataset presented can be used for comparisons to other ground- and satellite-based measurements and to petrological estimates of the SO_2 flux, and to initialise, for instance, climate model simulations, helping to better quantify the environmental and climatic impacts of volcanic SO_2 .

2 IASI SO₂ iterative retrieval scheme

30 IASI is an infrared Fourier transformer interferometer, on-board the Metop-A and Metop-B satellites. It measures in the spectral range 645-2760 cm⁻¹ with spectral sampling of 0.25 cm⁻¹ and has global coverage every 12 hours. The IASI data dataset used in this study was the level 1b-1c (apodized) dataset from the EUMETSAT and CEDA archive.

The details of the retrieval scheme are summarized briefly below. For more details see Carboni et al. (2012, 2016). An SO₂ retrieval is performed for all IASI pixels that present a positive result in the SO₂ detection scheme (Walker et al., 2011, 2012). The detection scheme uses all the channels in the range ν_3 band (1300-1410 cm⁻¹ (cm⁻¹) while the iterative retrieval uses all the channels in both spectral ranges ν_1 and ν_3 (1000-1200cm⁻¹ and 1300-1410 cm⁻¹). The strongest SO₂ band is the ν_3 band,

- 5 around 7.3 μ m, and is contained within a strong water vapour (H₂O) absorption band so it is not very sensitive to emission from the surface and lower atmosphere. However, above the lower atmosphere, this band contains valuable information on the vertical profile of SO₂. Fortunately, differences between the H₂O and SO₂ absorption spectra allow the signals from the two gases to be decoupled in high resolution measurements. The SO₂ ν_1 band around 8.7 μ m (1000 to 1200 cm⁻¹) is within an atmospheric window. This allows the radiation from the surface to reach the satellite from deep within the atmosphere enabling
- 10 the retrieval of SO_2 amount down to the surface.

The detection scheme is a linear retrieval with one free parameter, the column amount of SO_2 . In particular we assume the vertical distribution of SO_2 and the atmospheric vertical profiles (temperature and trace gases). We do not take into account negative thermal contrast so that regions with negative thermal contrast give zero (or negative) values of SO_2 column amount. In our scheme we consider detection 'positive' if the output of the linear retrieval is greater than a defined positive threshold

15 (0.49 effective DU, following Walker et al 2012). The detection limits for a standard atmosphere (with no thermal contrast) are estimate estimated to be: 17 DU for a SO₂ plume between 0-2 km, 3 DU between 2-4 km and 1.3 DU between 4-6 km (Walker et al., 2011). The detection scheme can miss part of an SO₂ plume under certain circumstances, such as low-altitude plumes, conditions of negative thermal contrast (i.e. where the surface is colder than the atmosphere), and where clouds are present above the SO₂ plume, masking the signal from the underlying atmosphere. Due to these uncertainties the estimated mass of

SO₂ in this paper should be regarded as a 'minimumminimum'. We perform the iterative retrieval for the pixels that give positive detection results. All the channels in the ranges 1000-1200 and 1300-1410 cm⁻¹ (the 7.3 and 8.7 μ m SO₂ bands) are simultaneously used in the iterative optimal estimation retrieval scheme to obtain the SO₂ amount, the altitude of the plume and the surface temperature. The scheme iteratively fits the forward model (simulations) with the measurements, through the error covariance matrix, to seek a minimum of a cost function.

- 25 The forward model is based on RTTOV (Saunders et al., 1999) extended to include SO₂ band around 8.7 μ m (1000 to 1200 cm⁻¹) is within an atmospheric window. This allows the radiation from the surface to reach the satellite from deep within the atmosphere enabling the retrieval of explicitly and uses ECMWF profiles interpolated to the measurement time and location. The error covariance matrix used is the 'global error covariance matrix' in Carboni et al. (2012). It is defined to represent the effects of atmospheric variability not represented in the forward model (FM), as well as instrument noise. This includes the
- 30 effects of cloud and trace gases which are not explicitly modelled. The matrix is constructed from differences between FM calculations (for clear-sky driven with ECMWF profiles) and actual IASI observations for a wide range of conditions, when we are confident that negligible amounts of SO_2 are present. Only quality-controlled pixels are considered; these are values where the minimization routine converges within 10 iterations, the SO_2 amount down to the surface.

is positive, the plume pressure is between 0 and 1100 mb and the cost function is less than 10. A comprehensive error budget 35 for every pixel is included in the retrieval. This is derived from an error covariance matrix that is based on the -free climatology of the differences between the IASI and forward modeled spectra. Rigorous error propagation, including the incorporation of forward model and forward model FM parameter error, is built into the system, providing quality control and comprehensive error estimates on the retrieval results. The IASI SO₂ retrieval is not affected by underlying cloud. If the SO₂ is within or below a cloud layer its signal will be masked and the retrieval will underestimate the SO₂ amount.

5 3 Temporal evolution of SO₂ mass and SO₂ vertical distribution

The retrieval results from the Metop-A orbits during the period from September 2014 to February 2015 and from 30° N to 90° N are combined (twice a day, i.e. morning and afternoon overpasses) to produce maps of retrieved SO₂ amount and altitude. Supplementary movie S1 shows the evolution of the plume for each day. The Holuhraun eruption is the main source of SO₂ over that period. Other minor sources include SO₂ emitted from intermittent volcanic activity on the Kamchatka peninsula, the Etna volcano (28th December 2014, 1st, 2nd and 21st January 2015), and anthropogenic SO₂ emissions from China/Beijing. Satellite

- 10 volcano (28th December 2014, 1st, 2nd and 21st January 2015), and anthropogenic SO_2 emissions from China/Beijing. Satellite observations at the pixel level do not provide sufficient information to distinguish between SO_2 from Holuhraun and SO_2 from other sources. For example, the elevated SO_2 near Beijing on 21st December 2014 appears to be from an anthropogenic source, but the elevated SO_2 in the same area on 31st December 2014 is from the Holuhraun eruption. In this study all the SO_2 measured from 30° N to 90° N between September 2014 and February 2015 is referred to as Holuhraun SO_2 .
- 15 Over the course of the six months the eruption plume dispersed across the Northern Hemisphere. Figure 1 shows the maximum SO₂ column amount retrieved during the six-month period and illustrates that SO₂ from the Holuhraun eruption was dispersed over large parts of the Northern Hemisphere including poleward of the Arctic circle. For the majority of the time the plume circulated around the pole and the northern regions (see animation in Supplementary Material Figure SXMovie S1), overpassing Scandinavia, Eastern Europe, Russia, Greenland and Canada several times. The plume overpassed Europe on mul-
- 20 tiple occasions, most often northern Europe (Schmidt et al., 2015; Ialongo et al., 2015; Zerefos et al., 2017; Steensen et al., 2016; Twigg et but also Italy (22nd-22nd October 2014) and Spain and reaching as far south as Morocco and Algeria (on 5th and 6th November 2014 respectively) and Greece/Macedonia/Albania (5th and 6th January 2015).

The mass present in the atmosphere for each IASI overpass was found by regridding the observations of column amount and plume altitudeinto a

- 25 IASI orbits are grouped into 12-hour intervals in order to have two maps, each day, of IASI retrieved SO₂ amount and altitude. These maps are gridded into 0.125° latitude /and longitude boxes following Carboni et al. (2016). The SO₂ mass time-series is obtained by summing the mass values of the regularly gridded map for each 12-hour 12-hour period. The same procedure has been used for the errors, this means that the sums of errors of grid boxes are considered as errors on the total masses (this could be an error overestimation but we cannot consider the usual errors in quadrature due to the possible presence
- 30 of systematic error, e.g. the errors are not independent). The time-series of SO_2 mass, together with the errors, are presented in the top plot of Figure 2. The largest SO_2 mass is highest found in September 2014 (up to 0.25 Tg) when the eruption was most powerful. The SO_2 mass decreases during October 2014 (with some peak values around 0.1 Tg) then increases around end of November/beginning December 2014 (up to 0.15 Tg). The SO_2 mass steadily declines during January and February 2015 as



Figure 1. Map showing the maximum of SO_2 column amount (in Dobson Units, DU) retrieved within the considered area from 30° N to 90° N (black rectangle) from September 2014 to February 2015.

the eruption comes to an end. There is no detection of \underline{a} SO₂ plume attached to the vent in the second half of February (and the SO₂ mass for this period, reaching a value up to 0.01 Tg, is from a non-Icelandic source).

The SO_2 mass present between two altitude levels was estimated using the method of Carboni et al. (2016) to produce the vertical distribution of SO_2 . In this study the vertical distribution of SO_2 was estimated every 12 hours from 0 and 10 km with a

- 5 vertical resolution of 0.5 km for all latitudes north of 30° N. Both the young emitted plume as well as the mature plume that had been transported around in the Northern Hemisphere for a few days are included in the distribution. The time-series of the SO₂ vertical distribution for the Holuhraun eruption is shown in Figure 3. The centre-of-mass of the plume closest to the vent can be used as a rough estimate for the injection height. Figure 3 shows the time-series of two datasets: (i) the vertical distribution and (ii) the altitude of the centre-of-mass of the SO₂ values within 500 km of the vent. The altitude of the centre-of-mass is
- 10 less than 4 km for the majority (96 %) of the measurements.



Figure 2. Time-series of the masses and emission fluxes of SO_2 mass as a function of day from $1st_1^{st}$ January 2014. Top The plot shows the IASI masses-mass (obtained from the retrieval) with error-bars error bars in black and the fitting of the flux retrieval (section 5) with the blue-green line. The bottom plot shows the retrieved flux time-series from IASI in black with grey error-bars Red circles and from IMO using DOAS in red (red bars show the errors, dotted bars orange line show the maximum-masses corrected for cloud presence and minimum values measured that day) their fitting of the retrieval.



Figure 3. SO_2 vertical distribution in km above sea level. The <u>colour colours</u> represents the mass of SO_2 , <u>dark-red dark red</u> represents values higher <u>than the colour-bar 0.03 Tg</u>. Every column of the plot is generated from an IASI map (one every 12 hrs; Figure 1 and supplementary files). The black diamonds show the altitude of the centre of mass (CM) computed with the IASI pixels within 500 km from the vent, the red and black dots show the <u>altitude from IMO-DOAS</u> measurements and other observations from Pfeffer et al. (2018).

4 Quantifying satellite-retrieval underestimation

Part of the SO_2 plume can be missed by this IASI scheme, and the derived SO_2 masses should be considered as minimum. The presence of low thermal contrast can prevent the detection of the SO_2 . While cloud below the plume should not be a problem for this scheme, the presence of cloud above the plume will smooth the spectral signature of SO_2 , causing underestimation of

5 the retrieved SO₂ amount (Carboni et al., 2012). At its most extreme this effect can completely erase the SO₂ signal (for a cloud layer with zero transmittance).

We estimate the percent of SO_2 mass missing due to cloud above the plume, as a function of cloud optical depth and altitude above the SO_2 plume, using the same simulations with a default atmosphere used in Fig 6 of Carboni et al. (2012). Using the European Space Agency cloud Climate Change Initiative dataset (Stengel et al., 2017, ESA cloud CCI) of Advanced Very

- 10 High Resolution Radiometer AVHRR (same platform as IASI, i.e. same local overpassing time) L3 monthly mean statistic, we computed:
 - Monthly mean histograms of frequency of cloud optical depth at 550 nm, τ , averaged over the globe. τ is not present in the cloud L3 database for locations without daylight (e.g. visible channels) and most of the Icelandic plume in the winter months is without daylight, as a consequence here we are assuming the global histogram of frequency of τ is valid over the plume region.
- 15 <u>the plume</u>
 - Monthly mean histograms of frequency of cloud altitude, averaged over the plume region (30° N to 90° N). Cloud altitude is available for all locations and during winter months.

(1)

We consider the measured mass M_{meas} to be the difference between true mass M_{corr} and the missing one M_{miss} :

 $\underline{M_{corr} - M_{miss} = M_{meas}}$

We can rewrite this as:

$$M_{corr}\left(1 - \frac{M_{miss}}{M_{corr}}\right) = M_{meas} \tag{2}$$

$$M_{corr} = M_{meas} \left(\frac{1}{1 - \frac{M_{miss}}{M_{corr}}}\right) \tag{3}$$

20 The term within the bracket on the right is the correction factor. We compute the correction factor, C, for every month of the eruption as a function of altitude, and apply it to the vertical distribution dataset.

$$M_{corr}(h) = M_{meas}(h) \cdot C(h) \tag{4}$$

With

$$C(h) = \frac{1}{(1 - Z(h))}$$
(5)

Where Z(h) is the fraction of SO₂ 'missed' in the measurements due to cloud above the plume. Z(h) is estimated as the product of probability of having cloud above altitude h, F(h), times the attenuation due to cloud, A,

5
$$Z(h) = F(h) \cdot A$$
 (6)

The probability of having cloud above *h* has been estimated from the ESA cloud CCI data for the region considered for the volcanic plume (latitude > 30° N) as the number of cloud retrievals above altitude *h* divided by number of observations.

Attenuation due to cloud (A) is the sum of the frequency of having a cloud with a cloud optical depth $f(\tau)$ times the attenuation due to a cloud with the same optical depth $a(\tau)$.

10
$$A = \sum_{\tau=0}^{n} f(\tau) a(\tau)$$
(7)

 $f(\tau)$ has been estimated using the monthly mean histogram of frequency of cloud optical depth, estimated over the globe. $a(\tau)$ has been estimated by running the SO₂ retrieval using, as IASI measurements, simulated spectra with water cloud above the plume, using the default atmosphere, and different optical depths τ . For optical depths bigger than 10 the attenuation is 1 (cloud is opaque and completely masks the SO₂ signal). Fig 4 shows the correction factor together with the SO₂ vertical

15 distribution obtained from the IASI retrieval and SO₂ vertical distribution corrected for underestimation due to cloud cover. In the following section the emission fluxes have been estimated with both the original SO₂ masses (obtained from IASI retrieval scheme explained in section 2) and the masses corrected by cloud cover (this section).

We estimate the missing SO_2 mass due to thermal contrast by comparison with OMI SO_2 (Theys et al., 2015) for the months of September and October 2014, as the OMI dataset does not cover all of the time period of the eruption due to the lack of

20 solar radiance during the winter. We compare, with visual inspection, the daily maps of IASI and OMI and identify the parts of plume missing from IASI detection (and consequently missing in the IASI retrieval). Table 1 presents the list of all areas identified and the SO₂ estimate from OMI (BIRA-IASB algorithm).

The total mass of SO_2 missed due to thermal contrast is estimated to be a few percent of the emission estimate by IASI. In particular the missing plume for the first 2 months, estimated with OMI 0-1 km, has a total mass of 0.08 Tg of SO_2 that

25 corresponds to 3% percent of the emission estimate by IASI (see section 5 for emission estimate). Cloud could be a problem also for OMI, for such low altitude SO₂, and OMI values could be off by a factor of 2-3, which will change the underestimation to 6-9% (instead of 3%).

5 Daily SO₂ fluxes

The time-series of SO_2 fluxes and the coefficient of an average exponential decay of SO_2 (the state vector **x**) are retrieved from 30 the time-series of SO_2 mass (the measurement vector **y**) using the optimal estimation scheme of Rodgers (2000). The solution



Figure 4. SO₂ vertical distribution in km above sea level. The colours represent the mass of SO₂, dark red represents values higher than the colour bar. Every column of the plot is generated from an IASI map (one every 12 hours). The top plot shows the data obtained from IASI maps. The middle plot is the top plot times the correction factor (to include SO₂ that statistically has been missed by the IASI measurements due to cloud above the plume). The bottom plot is the correction factor C(h).

Date	Max latitude	Min latitude	Max longitude	Min longitude	OMI SO ₂ for	OMI SO ₂ for
	[°_N]	[° N]	[°_ E]	[°_ E]	7 km heigh [kT])	0-1 km a.g.l. [kT])
20140901	<u>70</u>	<u>60</u>	-20	-36	7.25	<u>9.6</u>
20140915	<u>70</u>	75	-10	-20	9.5	11.1
20140915	75	<u>70</u>	<u>30</u>	0	21.5	33.7
20140929	<u>70</u>	.65	-15	-30	2	3
20140929	77	<u>.63</u>	-20	-40	12	26

(x, the vector of parameters that we want to retrieve) gives the most probable x given the measurements y y and the a priori knowledge x_a . The state vector x is found by minimizing the cost function:

$$\chi^{2} = [\mathbf{y} - \mathbf{F}(\mathbf{x})]^{T} \mathbf{S}_{e}^{-1} [\mathbf{y} - \mathbf{F}(\mathbf{x})] + [\mathbf{x} - \mathbf{x}_{a}]^{T} \mathbf{S}_{a}^{-1} [\mathbf{x} - \mathbf{x}_{a}]$$
(8)

where \mathbf{F} is the forward model (that simulates the measurement given the state vector), \mathbf{x}_a is the a priori value of the state vector and \mathbf{S}_e and \mathbf{S}_a are the measurement and a priori error covariance matrices. \mathbf{S}_e and \mathbf{S}_a are diagonal matrixes matrices

5 with the variances (square of errors) of y and x_a respectively as their diagonal elements. The a priori values used were 0.2±0.2 Tg/day for flux and 2±2 day for the e-folding time; these values reflect the variability of lifetime found in the literature for low tropospheric emission (Carn et al., 2016; Lee et al., 2011). To retrieve the time-series of SO₂ fluxes the state vector x was defined as follows. The first element of the state vector is the average SO₂ e-folding time (λ) for the period analysed; the following x_i elements are the average emission flux f_i between the two IASI estimates of SO₂ mass at time t_{i-1} and t_i:

$$\mathbf{x} = (\lambda, f_1, f_2, \dots, f_n) \tag{9}$$

To define the forward model \mathbf{F} we consider that the SO₂ mass m measured by the satellite is a function of time and that the mass decays proportionally to the mass itself, plus the addition of a source term of flux f. These terms give a first order differential equation:

$$\frac{dm}{dt} = -km + f \tag{10}$$

Assuming a constant flux f over the time interval Δt between two consecutive mass estimates m_i and m_{i-1} , the solution becomes:

$$m_i = m_{i-1}e^{-\frac{1}{\lambda}\Delta t} + f\lambda(1 - e^{-\frac{1}{\lambda}\Delta t}) \tag{11}$$

15 where λ (with $\lambda = 1/k$) is the average e-folding time. Equation 4-11 is the forward model $\mathbf{F}(\mathbf{x})$.

We used optimal estimation (OE) to fit the measured time-series of mass loading with a forward model that reproduces the time-series as a function of emission fluxes and SO₂ e-folding time. This forward model (eq. 11) is the inverse of equation 6 in

Theys et al. (2013), section 'Delta-M method'. Both equations derive from the solution of the same differential equation (eq. 10). In Theys et al. (2013) the fluxes are obtained assuming the e-folding time, here the fluxes and the averaged e-folding time and their uncertainties, are estimated simultaneously based on OE.

Figure 2 shows the fitting of the measurements \mathbf{y} with the forward model \mathbf{F} while Figure 5 shows the retrieved fluxes with 5 errors.



Figure 5. Time-series of emission fluxes of SO_2 . To facilitate the comparison with the flux estimates from Schmidt et al. (2015), the top plot presents values (for the first month only) of: the retrieved SO_2 flux time-series from IASI measured mass in black with grey error bars, the retrieved flux time-series from IASI 'corrected' mass in red with orange error bars and DOAS ground measurements from Pfeffer et al. (2018) in blue (blue bars show the errors, dotted bars show the maximum and minimum values measured that day). The bottom plot shows the 6-month time-series with the same colour coding.

The flux time-series follows a similar pattern to the SO_2 mass time-series, with higher highest values in September 2014. IASI determined fluxes are provided here with the ground-based measurement fluxes provided in parenthesis (described in section 5). The September daily average was 0.06 Tg/day (0.09) and the September daily maximum was 0.26 Tg/day on the 20th September (0.19). The October average flux was 0.023 Tg/day (0.083), with peak values of 0.15 and 0.12 Tg/day

- 5 on the 11th and 19th of October (0.10). The November average flux was 0.029 Tg/day (0.069). The flux increases in the second part of November to a maximum of 0.13 Tg/day on the 23rd of November (0.13). The estimates for December, January and February show decreasing flux with monthly average fluxes with monthly averages of 0.016, 0.006 and 0.005 Tg/day respectively (0.026, 0.028, 0.016). The monthly averages are lower than those measured by the ground-based measurements while the maximum daily averages for each month are generally higher. The UV ground-based measurements for the dark
- 10 months of December, January and February are sparse, with only 10 measurements over these 3 months. There was only one day with measurements at the beginning of December, and then 6 days with measurements in the second half of January and three days with measurements in February. The extrapolated flux from the ground-based measurements through December to the first half of January is consistent with the error bars from the IASI estimates. The differences in monthly means between the ground-based measurements and the IASI flux estimates in the sunny months are explainable by low values with large error.
- 15 The fluxes calculated for September 2014 are consistent with Schmidt et al. (2015) (e.g. up to $0.120 \ 0.12$ Tg/d during early September, 0.02-0.6 Tg/day between the 6th and 22nd of September, 0.06-0.120 Tg/day until the end of September).

The total mass of SO₂ emitted by the eruption is obtained by summing all the fluxes f_i, output by the retrieval and multiplying by the corresponding Δt_i. The error associated with the total mass emitted is obtained by adding in quadrature the errors δf_i multiplied by the time interval Δt_i. The maximum value of total mass emitted is obtained by summing all the fluxes
20 plus the uncertainty and the minimum value is obtained by summing all the fluxes less the uncertainty (negative values are set to zero). This gives a total mass of emitted SO₂ of 4.4±0.8 Tg with a maximum of 11.6 Tg and a minimum of 0.4 Tg. The

retrieved averaged e-folding time is 2.4 ± 0.6 days.

Note that in using an average SO_2 e-folding time for the entire eruption period, any variation of e-folding time will be interpreted as an inverse variation in the estimated flux, i.e when the 'realreal' e-folding time is higher than the retrieved one,

- the flux will be overestimated and vice versa. The flux uncertainties include the errors in flux due to the variation in e-folding time. The SO₂ lifetime can vary significantly on a daily basis mainly as a function of water vapour and solar irradiation. Also note that any e-folding time shorter then than the retrieved one can fit the measurements and give higher fluxes. Given these caveats the value of e-folding time $(2.4 \pm 0.6 \text{ days})$ is consistent with the mean lifetime of 2.0 days estimated for September 2014 (Schmidt et al., 2015). Their estimate was based on minimising the difference between the SO₂ amount from the NAME
 - 30 dispersion model and the IASI and OMI satellite measurements.

Figure 2-5 shows that higher values (peaks) of SO₂ flux often alternate with lower values (below 0.02 Tg/day) even during periods that have been identified as generally characterized by higher fluxes. This intermittent flux behaviour has important implications in terms of the estimate of total SO₂ emitted. Had a less sensitive instrument been used that only produced 'valid' measurements in correspondence with higher flux values (e.g. > 0.05 Tg/day), and had considered these fluxes as representative

of the period without valid measurements (i.e. period between two 'valid' measurements), this would result in very different (and higher/overestimated) estimated values of total emitted. An example of this is Gauthier et al. (2016) where they-

<u>Gauthier et al. (2016)</u> used TIR data from SEVIRI, on board on board the geostationary satellite Meteosat Second Generation (MSG), to retrieve an SO_2 mass time-series from 1 September 2014 to 25 November 2014. Their daily retrieved mass

- 5 values are lower compared to the IASI values here (reported here due to the smaller geographic area considered and possibly due to a smaller sensitivity or detection threshold of SEVIRI, nevertheless the lower sensitivity of SEVIRI. Nevertheless they estimate a total SO₂ emitted mass of 8.9 ± 0.3 Tg for the period September 2014 to November 2014, which is a factor of two higher than calculated hereIASI. Our understanding is that the Gauthier et al. (2016) scheme produced 'valid' measurements when the SO₂ loading increased with respect to the previous measurement. This leaves data gaps when the SO₂ measured
- 10 remains constant or decreases. The resulting data gaps were filled by linearly interpolating between two 'valid' measurements. This has a tendency to bias flux estimates in favour of increasing SO_2 loading. The dataset of Gauthier et al. (2016) contains several days with no valid flux estimates and, for these data gaps, the interpolation of valid measurements into the gaps could account for their discrepancies with our dataset. The ground-based measurements reported in Pfeffer et al. (2018) show an intermediate value of 7.3 Tg over this time interval.

15 6 Comparison with ground-based and near-source measurements

The conditions of the Holuhraun eruption are significantly different to eruptions investigated in previous studies using IASI (Carboni et al, 2016), because the plume from Holuhraun was confined to altitudes between the surface and 6 km at highnorthern latitudes and because the eruption took place during the autumn and winter months. As a result there is less radiance and low (or negative) thermal contrast between the surface and the first layer of the atmosphere. These conditions lead to lower

- 20 sensitivity for measurements in both the UV and TIR spectral range. Nevertheless a cross-comparison with other available measurements is informative when assessing our results. The following comparisons are an addition to previous comparison done with UV satellite and comparisons done between UV satellite measurements and a dispersion model (Schmidt et al., 2015) and balloon measurement measurements (Vignelles et al., 2016).
- First the IASI dataset is compared with ground-based Brewer measurements of the SO₂ column amount of the mature plume over Finlandand then. Successively the plume height is compared with near-source measurements in Iceland using: ground- and aircraft-based visual observations, web camera and NicAIR II infrared images, triangulation of scanning DOAS instruments, and the location of SO₂ peaks measured by DOAS traverses as reported in Pfeffer et al. (2018).

The Brewer ground measurements (Ialongo et al., 2015) were made at Sodankylä (67.42° N, 26.59° E). The SO₂ column amounts are routinely obtained from the direct solar irradiances at wavelengths of 306.3, 316.8 and 320.1 nm, by using the

30 same Brewer algorithm as for the ozone retrieval (Kerr et al., 1988). The method is based on the Lambert-Beer law, which describes the attenuation of the direct solar irradiance reaching the EarthEarth's surface at certain wavelengths due to the atmospheric constituents. In order to avoid the effects of stray light at short wavelengths, the measurements corresponding to large air mass values (after 14:20 UT) are not included. The SO₂ column amounts in Sodankylä are typically close to zero with

an estimated detection limit of about 1 DU. Ialongo et al. (2015) compared the SO_2 column amount values from the Brewer and OMI measurements in Sodankylä during September 2014 with 2014. The differences between OMI and Brewer retrievals were usually smaller than 2 DU.

The comparison here is performed by averaging all the IASI pixels that pass quality control, within 200 km of the ground
measurements. The Brewer instrument measures in the UV and thus only in daylight conditions. This means that only the first month of the eruption can be considered. Fig.Figure 6 shows the time-series of SO₂ column amount from both ground measurements and IASI as a function of time. All the 'plumeplume' episodes (with SO₂ amount larger than 2 DU) are consistent between the two datasets with the exception of 15th and 19th September where the plume only passes over the northern part of the 200 km circle in the IASI data and does not pass over the ground measurement station. This means that while IASI presents
high SO₂ measurements, elevated values are not observed in the Brewer measurements.

There are a few days of low (less the 2 DU) SO_2 reported by only one of the two instruments (IASI or Brewer), meaning that the detection limit of one of the instruments is not exceeded. This is consistent with the IASI minimum error for low altitude plume_plumes (2 DU for plumes below 2 km, Carboni et al., 2016) and with the Brewer minimum error (2 DU, Sellitto et al., 2017).

- 15 The ground-based measurements of eruption cloud top height were collated collected from multiple techniques including ground-ground and aircraft-based observations, web camera, ScanDOAS, MobileDOAS, and NICAIR II IR camera (Pfeffer et al., 2018). The ScanDOAS and MobileDOAS approaches, and IASI retrievals, provide the height of the center-of-mass centre-of-mass of the plume while the other techniques provide the height of the top of the plume. Fig 3 presents the groud-based ground-based and IASI altitudes together. In general, the IASI and ground-based altitudes agree that the altitudes varied mainly
- 20 between 1-3 km, however they do not agree particularly well on any specific day.

The time-series of the ground-based (Pfeffer et al., 2018) and IASI flux measurements estimates are presented in Figure 25. Within error, they generally agree. There are a few significant differences between the two datasets: two values in October/November 2014 and some values at the end of January and February. The ground-based measurements in November alternate between very high and very low values. On 5th November 2014, the ground-based value is significantly higher than

- 25 the IASI flux estimate for that day. Pfeffer et al. (2018) suggest the high values in November could be due to degassing from a continually replenishing lava lake contributing to the total gas in addition to the degassing from the magma being erupted. The plume altitude was less than 2 km on this day and under these conditions IASI values can be underestimates. The total mass emitted can be estimated, using the ground-based measurements, as the integral below the red line in Figure 2 (i.e. interpolating flux values). Even if the majority of fluxes are consistent with each other within the error estimate, the total mass calculated
- 30 by IASI (4.4 Tg) and IMO (9.6 Tg) differ differs by a factor of two. The 5th November discrepancy contributes significantly to this difference.



Figure 6. Time-series of the SO₂ column amount [DU] measurements at <u>Sodankylä-Sodankylä</u> as a function of day from the 1st September 2014. Black symbols are the Brewer measurements with error bars, red symbols are the mean and standard deviation of all the IASI measurements within 200 km, blue lines represent the maximum and minimum of the IASI measurements.

7 Conclusions

The first satellite-based SO_2 flux dataset of the full 2014-2015 Holuhraun eruption has been estimated using the IASI instruments. The dataset provides a flux estimate every 12 hours for the entire eruption. Thermal infrared spectrometers are the only satellite instruments that could follow the SO_2 plume around the Arctic in the absence of solar irradiation during the winter

- 5 months of the eruption. The low-altitude of the SO₂ plume and cold underlying surface reduce IASI's IASI sensitivity to SO₂, however the results compare resonably reasonably well with ground-based near and distal measurements. The observations show that the Holuhraun plume passed over large parts of the northern hemisphere Northern Hemisphere during the eruption. The time-series of vertical distribution the vertical distribution of SO₂ showed a low-altitude plume confined mainly within 0-6 km. The time-series of SO₂ masses showed a maximum of 0.25 Tg of atmospheric loading in September 2014. A new optimal
- 10 estimation scheme was developed to calculate daily SO₂ fluxes and e-folding time based on satellite-retrieved atmospheric SO₂ burdens. Application of the method gave estimates of SO₂ flux of up to 200 kt/day. The 'minimumminimum' total mass of SO₂ was calculated to be 4.4 ± 0.8 Tg and the average SO₂ e-folding time was found to be 2.4 ± 0.6 days.

By comparison with the OMI dataset during the summer months of the eruption we estimate that the SO_2 masses missed due to low thermal contrast are of the order of a few percent (3%) of the total emission. We estimated the SO_2 mass missed,

- 15 due to cloud above the SO₂ plume that is masking the signal, using AVHRR cloud CCI dataset monthly mean. Results show a correction factor increasing with decreasing altitude, from one (no underestimation of SO₂ masses) at the top of the cloud around 9 km, up to a factor two (we measure 50% of the 'true' mass) for plumes between 0-1 km. Applying this correction resulted in a total mass, emitted during the 6 months of the eruption, of 6.7 ± 0.4 Tg and little change in the average e-folding time (2.5 ± 0.7). The IASI flux data reported here are representative of every 12 hours and with no data-gaps. Other
- 20 emission estimates include the interpolation (or extrapolation) of fluxes where there are no measured data; this could produce discrepancies between estimates made using different methods.

Data availability. The SO₂ data presented in this paper are available from the corresponding author on request.

Competing interests. The authors declare that they have no conflict of interest.

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