The reviewers are thanked for their detailed and constructive reviews. As suggested by both reviewers, a substantial revision of the manuscript has been performed for exploring the impact on modeled far-range ground-level concentrations of 1) high spatial resolution model simulations (requiring an improvement of our computation capacities), 2) uncertainties on the source term and 3) the dynamics of the far-range planetary boundary layer.

These various tests allowed us to show the improvements in the modeled vertical distribution of the aged volcanic SO2 cloud reached with high-spatial resolution simulations. While variations in the altitude of SO2 injection at the source have a minor impact on far-range air quality modeling for this specific case-study, we show the key role played by the planetary boundary layer which is not accurately represented by state-of-the-art numerical weather prediction models.

Three new sections, including seven new figures, have consequently been added to the revised manuscript.

Answers to reviewers as well as changes made to the paper are detailed in the following in blue.

Anonymous Referee #1

Received and published: 13 April 2016

General comments

The paper by Boichu et al. presents a study on the far-range air pollution caused by the Badarbunga fissure eruption. The authors gather an extensive, and very useful, set of various type of measurements to complement and compare with modelling results. The paper is however rather descriptive and additional in depth evaluation of such an interesting dataset would be desirable. The authors encounter several problems in the modelling results that are not tackled. Although it is hard to tackle all of them, I would encourage the authors to suggest a roadmap on how to identify the main factors leading to a poor representation of the ground-level concentrations by the model. In addition, given that they have access to a full chemistry model, it would be worth to try to include SO2 chemistry in the simulations.

Given the significance of the event, both in terms of air quality and also on volcanic emission forecasts and potential impacts, the paper shall be revised and considered for publication once the main aspects stated before and below are are addressed.

Specific comments

Introduction:

- Page 2 Line 10-11 " Even so, the Bardabunga eruption only weakly disturbed air traffic..." is unnecessary since aviation implications are not the topic of the paper and they are mostly significant for ash-rich eruptions. If the authors want to still keep the reference to aviation, they may state that "Whereas the Bardarbunga eruption SO2 emissions were very large, but not constant, the ash emissions were limited and therefore no affectation in air traffic occurred, unlike on other occasions such as the Eyjafjallajokul eruption". It has been rephrased accordingly (page 2, line 25-26).

- Page 2 Line 12-13 "Nevertheless, Bardarbunga triggered a volcanogenic air pollution unprecedented in Europe. . . which necessitated locally exceptional civil protection measures". The sentence and posterior reference to Gisalson et al. 2015 (which only addresses the environmental stress in Iceland) is

misleading since it reads as if exceptional civil protection measures were taken also in areas of Europe other than Iceland, please rephrase. It has been rephrased (page 2, line 28).

- Page 2 Line 17 "The Bardarbunga cloud travelled most often . . .. toward high latitudes". Please add reference, even if it is, for instance, Figure 4 of the manuscript. References to Fig. 4 (illustrating IASI SO2 altitude) and McCoy et al. (2015) GRL paper have been added (page 2, line 34).

- Page 2 Line 18 "peculiar meteorological conditions". They were not that peculiar given that, for instance, the Eyja event suffered similar transport conditions transporting the ash plume rapidly over mainland Europe. I would suggest "favourable conditions" this sentence was indeed confusing, it has been modified accordingly (page 2, line 34)

- Page 2 Lines 25 onwards until the end of the paragraph " Here, we use a wealth . . .. " is ambitious given that the paper is so far more descriptive and does not go in depth into the characterization of SO2, the derived sulfates and the dynamics of the ABL leading to such unusual concentrations at ground level. Please revisit this sentence after addressing the comments here presented and rephrase if needed. In addition, although the comparisons are indeed quantitatively, they require further analysis and better description in the text to be stated as it is now.

We disagree with this comment, mentioning that this article is essentially descriptive, which seems to us very unfair.

Indeed, we first document this event by gathering and exploiting a large panel of volcanic SO2 observations from space (IASI, OMI), from ground-based remote sensing instruments (UV MAX-DOAS) and from in-situ ground-level air quality measurements at various locations. Moreover, we also gathered a vast panel of complementary observations of aerosols (sunphotometric, lidar and ground-level in-situ particulate measurement). The synergistic analysis of these various observations, using state-of-the-art retrieval algorithms, allowed us to provide a detailed characterisation of sulfate aerosols in a tropospheric volcanic cloud including timeseries of far-range vertical distribution, aerosol optical depth, volume size distribution with time and single scattering albedo. To our knowledge, such a detailed characterisation is rarely, if not never, available for a tropospheric plume compared to stratospheric plumes whose

lifetime is considerably longer rendering their observtion facilitated.

The combined analysis of both SO2 and sulfate aerosols allowed us to accurately describe the large-scale dispersal of the Bardarbunga cloud, its descent down to the lower troposphere and its arrival at the ground level.

Thanks to this detailed description of the volcanic cloud behaviour and additional observations of the far-range dynamics of the planteary boundary layer, a strong modeling effort was made to thoroughly test whether our modeling capacities are currently sufficient to simulate with accuracy this large-scale volcanogenic event of air pollution. After exploring various directions, our modeling study allowed therefore to show the various sources of difficulty encountered to reproduce correctly the magnitude of a volcanogenic pollution episode (spatial resolution of model simulations, uncertainty on source term, PBL dynamics). More precisely, this article (in its revised form) points out the key role played by the PBL dynamics and the limits of current state-of-the-art numerical weather prediction models for modeling it with sufficient accuracy for our case-study.

As such, we think that our paper brings indeed a wealth of complementary observations of SO2, sulfate aerosols and PBL dynamics. Such a panel of observations allowed us to perform a substantial modeling exercise and to highlight clearly the barriers which still need to be overcome in the coming years to accurately simulate and forecast such a volcanic vent in the coming future.

Methodology:- Page 3 Line 8 “Given the low injection height” needs a reference. Reference to Fig. 1 (previously Fig. 4 in the ACPD version), which shows an altitude of SO2 below 4-5 km near Iceland at the end of September 2014 and confirms the low injection height mentioned in the manuscript, has been added (page 3, line 20)

-Page 3 Line 10 “The center of mass of the SO2 cloud is assumed to be within the PBL”. What is this important statement based upon?

The choice of the OMI SO2 product, between here PBL or TRL (lower troposphere) products which are associated respectively to a center of mass altitude of 0.9 and 2.5 km, has to be made according to independent information on the altitude of the volcanic cloud which are scarce for the

Bardarbunga eruption.

The chemistry-transport model allows for reproducing far-range ground-level concentrations assuming that a part of the emissions are injected at 1 km. Moreoever, IASI shows that the altitude of the SO2 cloud near Iceland is below 4-5 km.

In this context, we tested both PBL and TRL products. The best agreement with model simulations (in terms of extent of the volcanic cloud) is reached with the OMI PBL product which is consequently chosen.

- Page 3 Line 20 The reference to the figure 1 should be complemented with additional explanation of the figure in the text. How this figure relates to the event the authors are examining? Are they suggesting that these low tropospheric aerosols are partly due to the event? What is the relation of the figure 1 with the topic of the paper?

The present reference to this figure in the text is indeed unclear as it seems to have been misunderstood. The text (page 4, lines 2-4) and caption of Figure 2 (previously Fig. 1) have been modified for clarification. This figure is useful to show that, while lidar observations are used to detect any kind of atmospheric particles, meteorological clouds can be clearly distinguished from aerosols (partly of volcanic origin here) in our case-study using adapted retrieval algorithms. Indeed, meteorological clouds evolve at a higher altitude that the aerosols that interest us lying at a low altitude (below 1.2 km).

- Page 3 Line 4 As in the previous comment, reference to figure 2. The figure is presented but all the information one can extract from the figure is not written in the text. Please do so and clearly state how the figure relates with the influence of the volcanic eruption.

For clarification, additional explanations on the exploitation of these data for characterizing volcanic aerosols have been added to the revised manuscript (page 4, lines 10-19).

- Page 4 line 10 of “Chemistry-transport model”. The authors state that the conversion from SO2 to SO4 is not implemented to avoid uncontrolled influence of uncertainties on the numerous factors governing this process in a volcanic cloud. It is unfortunate that the authors decided not to study the

conversions since then the comparison with the aerosol measurements would have been more interesting. Given the characteristics of the eruption, with such a low height emission and transport, the conversions from SO2 to SO4 may be significant and one would hope that the CTM would at least reflect part of it. Have the authors at least tried to include the conversions? Given that the authors use a CTM, I would encourage them to add discussions on this and, if possible, an additional test with the conversion activated. Otherwise one may wonder why using this model and not something closer to a Lagrangian particle dispersion model.

The difficulty highlighted in our paper published in ACPD was to understand the reason why we missed entirely the second peak of ground-level SO2 concentration at all the studied monitoring stations. Including the conversion of SO2 gas to sulfate aerosols would not help to solve this problem. Indeed, the current absence of conversion leads only to an over-estimation of far-range SO2 concentrations and, subsequently, an overestimation of the SO2 emission flux (added page 5, lines 20-21). However, we have elucidated the reason for this problem in the revised manuscript by exploring the impact on ground-level concentrations of runninng model simulations at high spatial resolution, of the PBL dynamics, and of the source term (adding 3 new sections and 6 new figures).

As indicated by the reviewer, sulphates aerosols of volcanogenic origin may be important during such eruptions. Efforts will be made in the near future to implement this SO2 conversion into the chimere model. However, we think that a detailed validation study is also required prior to using the conversion scheme blindly. Taking advantage of the various observations that we gathered in this study and of the detailed characterization of sulfate aerosols that we performed, it represents the scope of another study.

- Page 5 line 17. WRF can work using different PBL schemes. Is there any reason for using YSU in particular? Where there some sensitivity tests behind that suggested this one to be the one giving the best results? Given that the evolution of the PBL is crucial in this event to understand the ground level concentrations, more details on additional sensitivity tests, if done, would be useful and help understand the influence of this very important parameter in the final ground-level concentrations. Although not all the potential tests should be presented, for the sake of keeping the manuscript short, any insight

in significant parameters is valuable.

YSU parameterization scheme has been used in initial simulations as it is the most widely used scheme in the WRF model. We tested in the revised manuscript two additional PBL parameterization schemes (ACM2 and MYNN3) and showed the key role played by the PBL dynamics to accurately simulate far-range ground-level SO2 concentrations in our case-study.

For this purpose, a new section (Section 4.4) was added including three new figures (Figures 13, 14 and 15). It shows how the PBL scheme controls the timing and amount of capture of the overlying volcanic SO2 cloud by the boundary layer, and subsequently the timing and magnitude of increase of the SO2 concentration at ground-level a few hours later. A large variability (up to ten-fold) of ground-level SO2 concentrations according to the chosen PBL scheme is highlighted.

Even if the ACM2 scheme provides the best fit to observations, none of the PBL schemes allows for rigorously modeling the second SO2 peak concentration with correct timing and intensity. We show that this difficulty likely results from the inaccuracy of the modeled PBL height time series. Indeed, this latter presents marked differences with observations retrieved from lidar observations, with a large underestimation of the modeled PBL height especially during mornings and evenings.

- Page 5 Line 25 “inception time” what does this mean in this context? This means 'time of release'. This has been modified in the text for clarification.

- Page 5 Line 25 onwards: as for what I understand, the authors modified the injection height and times trying to match as much as possible the satellite data keeping a gaussian profile. Did they do this automatically or by simple visual inspection? It would be useful to know. It is also important to note that, the coarse assumptions in the source term make an accurate evaluation difficult. It would be good to highlight this in the conclusions section and state that the aim of the paper was not to make an estimate of the source term but to try to accommodate a simple source term that would represent the main features for this far-range study. A plot with the source term (injection height, times, vertical profiles) used in the modelling would be very useful to accompany figure 4 and would help the reader visualise the simulation.

The objective of this study is not to provide an accurate description of source emissions but to show that a simple source term is indeed able to reproduce main features of a far-range volcanogenic air pollution event. For this reason, the best fit to satellite and ground-level SO2 concentrations was evaluated by visual inspection. This has been now better mentioned in the text (page 6 line 1).

The reconstruction of detailed emission timeseries for the whole duration of the eruption using inverse modeling tools, will be the subject of another paper in preparation.

An additional figure (Fig. 5), representing the simple source term used to initialize chemistry-transport simulations of the Bardarbunga SO2 cloud dispersal toward Europe, has been included in the revised manuscript.

- I would suggest also more description of Figure 4. For instance, we can clearly see from the derived IASI heights that for lower latitudes the heights are constrained to heights mostly below 8km. In addition, over many areas, example 20/09/2014 UK, the cloud is constrained below approximately 5 km a.s.l which will of course favour potential plume ground-touching.

A more thorough description of IASI images of the SO2 altitude was indeed lacking in the ACPD paper, as they allow for explaining a lot of features (such as the absence of the traces of SO2 over mainland Europe on 22 Sept) which are not reproduced by the model as they are associated to emissions released and then transported at high altitude toward regions (like northern Greenland, Fenno-Scandinavia) which are out of our domain of interest here (i.e. Western Europe). These descriptions have been added to Section 3.1 (page 6, lines 13-24).

Results:

As previously stated, if possible, it would be good to include a simulation that accounts for the SO2 conversions to sulfates. Answered earlier

- Section 3.1 title, Large scale SO2 dispersal from Iceland toward Europe does not read nice. I would suggest Large scale transport of SO2 towards Europe : title has been modified

- When looking at Figure 5, one has at least some doubts about the transport

towards the Atlantic ocean of the Wave 1 since OMI show some traces that could actually be wave 1 transported further into mainland Europe. What is the opinion of the authors on this?

An animation has been added to the revised manuscript (in the Supplementary Material), which shows more clearly than maps how Wave 1 is transported toward the Atlantic Ocean. According to IASI SO2 altitudes (Fig. 5), the SO2 traces detected by OMI on 22 Sept further above mainland Europe lie at a higher altitude (between 8 and 10 km asl) than Wave 1 (lying at an altitude below 4 km). According to IASI images of the SO2 altitude for the previous days, these traces are likely associated to a part of the the SO2 emissions released at a high altitude (above 8 km), transported first toward the Pole then dispersed over Fenno-Scandinavia. As stated in Section 2.2, these parcels of emissions, which do not travel toward Western Europe, are not taken into account in the modeled source-term.

Some explanations on these parcels of the volcanic SO2 cloud have been added to the revised manuscript (page 6, lines 13-24).

- Page 5 line 15-16, have the authors tried to gather data from the Scandinavia region to further assess the model behaviour in this region?

We have not tried to gather data in other regions as we could only gather for France (and nearby Belgium) a wealth of complementary observations on SO2, sulfate aerosols and PBL dynamics, the latter being particularly crucial to understand the key role played by the PBL and the source of discrepancies between model and observed far-range ground-level concentrations.

Moreover, according to us, the simple 2-wave behaviour of our french case-study, is an exceptional opportunity to assess our current modeling capacities.

Accordingly, adding additional observations is obviously of interest. Another study is underway to compare Eulerian vs. Lagrangian model approaches, as well as Era Interim/ECMWF vs. NCEP meteorological forcings. It will include many more stations in France, UK, Netherlands and Scandinavia.

- Figure 6 c: why are the ground-level concentrations of SO2 and particles de-phased with particle concentrations peaking several hours after the passage of the SO2 plume? Whereas the text states there is coexistence of SO2 and

sulfates, we see a delay in the peaking particle concentrations. We see this behaviour both for the first and second waves.

Indeed, SO2 and sulfate aerosols are more generally co-existent, e.g. a study of the Etna volcanic cloud by Boichu et al., ACP, 2015. Nevertheless, we need to mention that we do not have exhaustive observations to document thi co-existence as the signature of tropospheric sulfate aerosols of volcanic origin is difficult to isolate from the signature of co-existent of aerosols/particles of other type, especially in highly-polluted regions.

ACSM (aerosol chemical spectiation monitor) observations, which are not presented in this paper but included in another study in preparation, allow for characterizing the detailed chemistry of PM1 particles. They indicate that Bardarbunga SO2 and sulfate aerosols at ground-level are co-existent and follow a similar temporal pattern.

According to us, this shift between ground SO2 and particulate matter peaks consequently indicates the ground-level pollution by particles which are not only of volcanic origin. This result demonstrates the difficulty to rely only on ground-level PM observations, which do not provide information on the chemical signature of sampled aerosols, to identify and isolate the signature of aerosols of volcanic origin from co-existing aerosols of various possible origins especially at ground-level in a urban context which can be highly polluted.

Also, seeing the plots, it would be good to add a discussion of the PBL evolution and how this is may be influencing the concentrations at ground-level.

As mentioned above, an extensive exploration of the influence of various PBL parametererization schemes on far-range ground-level SO2 concentrations has been developed in the revised version of the paper (new Section 4.4) and allowed to show the crucial role played by the PBL dynamics to accurately model large-scale events of volcanogenic air pollution.

- Page 7 line 25 “Interestingly...”. Why are the authors surprised about the two cities following a similar pattern? In sections before, the authors describe the transport patterns by explaining two waves coming towards Europe. This,

therefore, makes it evident that the temporal patterns of the two locations may undergo a similar signal pattern. And actually the authors stated this right after the “Interestingly. . . “ sentence. I would rephrase it and start with “ As observed from space and reproduced by the CTM, two waves. . .. This is also seen in the measured ground-level concentrations at ...”

These results seem not to be correclty presented in the ACPD version of the paper as the presence of a two-wave pattern is currently a result of our study combining model and observations, which allows for linking spatial to ground-based air quality observations thanks to model simulations.

We were indeed surprised to highlight such an interesting simple pattern, that we could follow in time progressing from the North to Central France at various ground monitoring stations. Compared to other studies performed in other regions (UK, Netherlands, Scandinavia) where air quality data seem to present a more ‘chaotic’ or ‘disordered’ behaviour (e.g. Ialongo et al., 2015; Schmidt et al., 2015), this french case-study seems consequently particularly interesting, as a kind of ‘textbook case’, for testing our current modeling capacities.

Text has been modified to clarify these points (page 8, lines 1-4).

- The authors state that the model fails to represent the second wave. Looking at the magnitude of the model at the first wave I am wondering whether what actually happens is that the model is maybe too fast and representing the second wave too early. Do the authors have any comments in this regard? Or, if not, do the authors have any suggestion on why the much significant peak is not at all captured by the model? Is it a transport problem? A mixing problem? A combination? Is it due to the assumptions in the source term? Given the discussion further on, it seems that the authors are, understandably, concerned about the representation of the PBL height. Have the authors made any tests in this regard? Also, as stated before, different PBL schemes in WRF can create different output. It would be good to have a clearer opinion of the authors on what factor they consider may be influencing most the poor model performance when representing the ground level concentrations and how would they approach a study to discern what is the main effect and how to compensate it (for example, as they have already suggested, increasing the resolution of the CTM and NWP calculations)

In the submitted ACPD paper, we suggested that the missing 2nd peak in ground-level concentrations resulted from an incorrect description of the vertical distribution of the volcanic cloud flying other the various monitoring ground stations. In this initial version of the article, we proposed as a future workplan to first develop simulations at a higher spatial resolution which could help to more accurately describe the long-range transport/dispersal of the volcanic cloud and its descent then its capture by the far-range boundary layer. We also suggested to explore the impact of the modeled PBL dynamics and the lack of a detailed knowledge of the source term.

In the revised manuscript, we explored these three hypotheses. For this purpose, we added three new sections (Sections 4.2, 4.3 and 4.4) which include six new figures (Figures 10 to 15).

First, we show that large improvements in air quality modeling are reached with simulations at higher horizontal resolution (Section 4.2). Initial simulations in the ACPD version were run using one single domain with a 25 km x 25 km horizontal grid resolution. After an upgrade of our computation capacities, we run, for the revised manuscript, simulations on two nested horizontal domains : the largest domain (from north Greenland down to Spain) with a coarse resolution of 22 km x 22 km and the narrower domain (large nevertheless from Norway down to Central France) with a finer resolution of of 7.3 km x 7.3 km.

We show how these high spatial resolution simulations allow for more accurately describe the vertical distribution of the far-range Bardarbunga cloud and its descent over France with time.

Despite clear improvements, discrepancies between modeled and observed SO2 concentrations at ground level remain.

Secondly, we introduce a variation in the flux and altitude of source emissions. For our specific case and period of study, we show that the source term plays a minor role and cannot solve the disagreement (Section 4.3).

Thirdly, as already mentioned above, we explore the impact of the PBL dynamics and show its key role to accurately model far-range voplcanogenic air pollution episode (Section 4.4). However, among the various PBL parameterization schemes that we tested, none of them allows for correctly

reproducing the right timing and intensity of capture of the overlying Bardarbunga cloud by the far-range boundary layer and subsequently the ground-level pollution episode. The substantial differences between modeled and observed (using lidar measurements) PBL height timeseries highlight the current limit of state-of-the-art mesoscale meterological models to solve this matter.

Interactive comment on "Tracking far-range air pollution induced by the 2014–15 Bárdarbunga fissure eruption (Iceland)" *by* Marie Boichu et al.

Anonymous Referee #2

Received and published: 18 April 2016

General Comments

The paper 'Tracking far-range air pollution induced by the 2014–15 Bárdarbunga fis- sure eruption (Iceland)' describes a modelling exercise based on this particular erup- tion complemented by a large range of measurements. The paper is well written and well-structured and does a good job of highlighting notable and challenging aspects as- sociated with this work, although there are a number of issues relating to the modelling aspect of the work that would need to be addressed before publication.

Specific comments

Discrepancies between models and observations are discussed and a number of reasons have been assigned to this. Possible explanations for these differences include:

Flux emission and altitude of injection 'This discrepancy can result from a limited knowledge of SO2 emission parameters (flux and altitude of injection) which initialize the chemistry-transport model.' It is also stated that 'Inception time and altitude of emissions are found by trial and error so as to reproduce first-order features of satellite and ground-level SO2 observations'. As the authors state model inversions can help with the refinement of this source term and help to further understand this discrepancy. This is clearly outside the scope of the work presented here although other possible reasons for the discrepancy may warrant further clarification. I think a more full discussion of SO2 oxidation and its possible contribution of the discrepancy should be included (after all it is a CTM). It is stated 'However, the conversion of SO2 to sulphate aerosols is not implemented in this study to

avoid uncontrolled influence of uncertainties on the numerous factors governing this process in a volcanic cloud'. This is a reasonable approach although ground based measurements of sulphate aerosols suggest a fairly significant conversion which is not reflected in the source term. The inclusion of these interactions in future model iterations would clearly represent an improvement.

Exploration of the impact of uncertainties in the source term has been made in the revised version by testing various altitude of injection of SO2 emissions (from 3 to 7 km a.s.l.) and various emission flux values. We showed that in our specific case and time period of study, the source term plays a minor role on the far-range SO2 concentration at ground-level (new Section 4.3), compared to the spatial resolution of simulations (new Section 4.2) and the PBL dynamics (new Section 4.4).

Moreover, the objective of our paper is not to provide a detailed source-term for the Bardarbunga eruption. Reconstructing the Bardarbunga source term over the course of the whole eruption is nevertheless the goal of another paper in preparation developing inverse modeling procedures.

Our objective in the article here is to reproduce first order features of the far-range air pollution event triggered by this eruption using a simple source term. Hence, we showed that simulations at low spatial resolution with a simple source term do not allow for correctly representing this far-range pollution as the second large peak of SO2 concentration recorded at all monitoring stations is entirely missed.

The absence of conversion of SO2 to sulfate aerosols would not help to solve this problem. Indeed, this configuration only leads to an overestimation of the far-range SO2 abundance (as more SO2 should disappear at distance from the source by their conversion to sulfate which is currently not taken into account). Consequently, this process cannot help to reconciliate observations and model, where the modeled second peak of concentration is already substantially under-estimated (even completely missing).

Subsequently, this absence of conversion leads also to an over-estimation of the SO2 source term. However, reconstructing in detail the Bardarbunga source term in late September 2014 is not the goal of this paper. In addition, we think that validating the modeling of SO2 conversion to sulfate aerosols in

a tropospheric volcanic plume, with relevant observations of sulfate, is of crucial importance. Identifying and isolating the signature of sulfate aerosols of volcanic origin in a mix with aerosols of various types, as is commonly the case in polluted tropospheric regions, is also a challenge.

Therefore, taking advantage of the panel of volcanic sulfate observations gathered in this paper and their detailed characterization (time series of vertical distribution, aerosol optical depth, volume size distribution and single scattering albedo) retrieved here using state-of-the art algorithms, as well as additional observations not included in this article, we aim at developing a thorough validation of sulfur oxidation in a tropospheric volcanic plume using our chemistry-transport model in another article.

Observations of the boundary layer heights compared to model simulations show a very large underestimation with the largest differences being observed at night time. The authors suggest that this is a ubiquitous feature of WRF. I would recommend confirming the influence of the boundary layer parameterisations by running WRF simulations using a number of parameterisations. This would confirm the influence of boundary layer height on the results presented here and may help to understand its contribution to model/observation mismatch.

This point has also been raised by reviewer 1. For more details, please refer to the detailed answer made above. In a few words, we have indeed showed the key role played on the representation of far-range ground-level concentrations by the dynamics of the PBL (new Section 4.4). To do so, we have run multiple simulations for testing three PBL parameterization schemes (the most widely used YSU, but also ACM2 and MYNN3 schemes). These investigations showed that the ACM2 delivers the best fit to ground-level concentrations but some discrepancies remain on the timing (late of a few hours) and intensity (underevaluated by a factor of 3) of the second peak of concentration observed at far-range monitoring stations. The comparison of modeled PBL height timeseries with observations retrieved from lidar measurements allowed us to show that the inaccuracy in PBL model representation explains this shortcoming. Hence, this case-study illustrates how we reach here the limits of current state-of-the-art numerical weather prediction model, as the PBL dynamics represents one of the most challenging modeling task.

A specific section (Section 4.4) to develop this point, including 3 new figures, has been added to the revised version of the manuscript.

It is suggested that higher model resolution (temporal and spatial) may help elucidate further the source of observation/model differences and this has both further time and computational costs. This is a perfectly reasonable argument. However I do not think it would be not beyond the scope of this study to perform some test simulations at a higher resolution in order to shed light on this point.

This point has also been raised by reviewer 1. For more details, please refer to the detailed answer made above. In a few words, we have run simulations at higher horizontal and vertical resolutions which required first to improve our computation capacities (Section 4.2). Such new simulations are indeed highly ressource consuming as we run simulations with 2 nested horizontal grids. The narrower domain, extending nevertheless on a large region from Norway down to Central France, has a finer resolution of 7.3 km x 7.3 km. We showed how the higher horizontal resolution allowed for improving the long-range transport/dispersal of the volcanic cloud and the far-range vertical distribution of the volcanic SO2 cloud. Hence, the descent of the Bardarbunga SO2 cloud over France occurs earlier and at a higher speed than modeled with lower spatial resolution simulations. The volcanic cloud also touches now the ground on 22 Sept, which was not the case with initial simulations. Consecutive to these improvements, the model simulates a second peak in SO2 concentration, which was entirely missing with initial simulations at low resolution as the modeled Bardarbunga cloud was not descending down to the ground on 22 Sept.

A specific section (Section 4.2) to develop this point, including 3 new figures (Figures 10, 11 and 12), has been added to the revised version of the manuscript.

In short I would suggest that perhaps a small effort in performing some simulations using a selection of boundary later parameterisations in WRF. Higher resolution simulations, if possible, would also help to strengthen (or at least clarify) some of the ideas presented here. A more complete discussion of the SO2 oxidation should be also included.

The above-mentioned explorations of the impact of high-spatial resolution

simulations, PBL dynamics and source term have allowed to hierarchize the factors responsible for the discrepancies obtained between modeled and observed far-range ground-level concentrations. These investigations, developped in three new sections in the revised manuscript (Sections 4.2, 4.3 and 4.4), show that the source of model shortcomings mainly result from an inaccurate modeling of the PBL dynamics.

Exploring some other locations to confirm the model performance in other regions and add more credence to discussion and conclusions should be considered.

As mentioned earlier, the french case-study is especially relevant as we were able to highlight a regular pattern with two large peaks in SO2 concentration recorded at all ground-monitoring stations (and only separated by a time shift of a few hours). From a combined analysis of osbervations and model simulations, we showed that this specific behaviour results from the arrival of two volcanic SO2 waves over France. Compared to air quality observations published for other stations (in UK, Netherlands and Scandinavia) which present a more disordered behaviour, the regular dynamics of the french case-study represents a kind of textbook case, which seems to us particularly interesting for testing the accuracy of our model simulations.

Moreover, in addition to the exploitation of a large panel of SO2 and sulfate observations, we also have, for this french case-study, simultaneous information on the PBL dynamics retrieved from lidar observations. To our knowledge, such a rich panel of information is not available elsewhere and was crucial to understand and conclude on the crucial role played by the far-range PBL dynamics for robustly simulating volcanogenic air pollution events, and the current limitations of state-of-the-art NWP models for accurately modeling its dynamics.

Perhaps the authors might outline a possible framework for a set of simulations that might elucidate these uncertainties. The conclusion reiterates the issue surrounding the boundary layer in the model but this should be contextualised within the framework of the other possible reasons for model-observation mismatch.

Technical corrections

Page 1 Line 1 ‘has emitted’- is ‘has’ necessary? removed

Page 1 Line 3 ‘chemistry – transport’ –model should be included after this for clarification added

Page 2 Line 13’ triggered a volcanogenic air pollution unprecedented’. Either ‘a’ should be removed or a descriptor after ‘air pollution’ should be included. corrected

Page 4 Line 10 Do you need three references from the same author here? First and last references were only kept

Page 4 Line 12 This sentence regarding the omission of the SO2 chemistry could be improved. This will clearly lead to large uncertainties when comparing to SO2 mixing ratios. The measurements of the sulphate aerosols provide some information regarding the magnitude of the conversion process and should be included here

As explained above, not considering the conversion of SO2 to sulfate aerosols will lead to an over-estimation of SO2 source emissions to fit far-range SO2 abundance. This is not an issue as the objective of our study is not to provide an accurate but a first-order estimation of the Bardarbunga source in order to show that it can reproduce first-order features of the far-range SO2 column load and ground-level concentrations. These explanations have been added to the text of the revised manuscript (page 5, lines 19-21).

Page 5 Line 14 What was the spin time up on the WRF simulations?

The spin time up of WRF simulations is of five days. Added page 5, line 23.

Section 2.2 Line 24 What is the justification for choosing a Gaussian profile?

This choice has been made to point out the impact of a specific altitude of injection.

Section 3.1 Line 10 perhaps ‘hitting’ could be replaced with reaching replaced

Figures

Figure 1 – It is hard to see how figure 1 is directly related to the text provided.

As reviewer 1 made the same remark, the answer is already developed above. Text (page 4, lines 10-19) and caption of Fig. 2 (previously Fig. 1) have been consequently modified for clarifying this point.

Figure 6c- Why might there a time shift between gas and aerosol?

As developed for reviewer 1, ACSM observations (not included in this paper) providing a full chemistry description of PM1 component, shows that SO2 concentration is clearly correlated in time with sulfate aerosol concentration. From our point of view, this shift rather points out the mixing in the boundary layer of aerosols of various origins, not only volcanic. Contrary to SO2 which is a clear and unambiguous volcanic indicator, this shift shows the difficulty to extract the aerosol component of purely volcanic origin. This objective cannot be reached using only ground-level particulate matter observations. It requires more diverse measurements, such as photometric data explored in this paper which allow for fully characterizing sulfate aerosol optical depth, size distribution and single scattering albedo using state-of-the-art retrieval algorithms, in order to identify the signature of volcanic sulfate aerosols.

Figure 9 – What would be an estimate of the uncertainty on the model boundary layer simulation?

As already developed in answers to reviewer 1, many additional simulations for testing the impact of various WRF PBL parameterization schemes allowed us to estimate the associated uncertainty on modeled ground-level SO2 concentrations at a far distance from the volcano (see new Fig. 13 and 14). It also showed the limitations encountered with state-of-the-art NWP models as not any of these various PBL schemes is able to correctly reproduce the expected dynamics of the PBL as retrieved from lidar measurements (new Fig. 15).

Discussion - In the discussion the phrase ‘finding optimum configuration’ is used. This is something that could be undertaken or considered with the boundary layer parameterisation within WRF. This work would certainly strengthen some of the conclusions presented in this work.

As explained above, the impact on modeled far-range ground-level SO2 concentrations of both simulations performed at a higher spatial resolution (new section 4.2) and of various PBL WRF parametrisation schemes (new

section 4.4) has been developed in the revised manuscript.

Current challenges in modeling far-range air pollution induced by the 2014–15 Bárðarbunga fissure eruption (Iceland)

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Abstract. The 2014–15 Holuhraun lava-flood eruption of Bárðarbunga volcano (Iceland) emitted prodigious amounts of sulfur dioxide into the atmosphere. This eruption caused a large-scale episode of air pollution throughout Western Europe in September 2014, the first event of this magnitude recorded in the modern era. We gathered chemistry-transport simulations and a wealth of complementary observations from satellite sensors (OMI, IASI), ground-based remote sensing (lidar, sunphotometry, differential optical absorption spectroscopy) and ground-level air quality monitoring networks to characterize both the spatial-temporal distributions of volcanic SO_2 and sulfate aerosols as well as the dynamics of the planetary boundary layer. Time variations of dynamical and microphysical properties of sulfate aerosols in the aged low-tropospheric volcanic cloud, including loading, vertical distribution, size distribution and single scattering albedo, are provided. Retrospective chemistry-transport simulations at low horizontal resolution (25 km × 25 km) capture the correct temporal dynamics of this far-range air pollution event but fail to reproduce the correct magnitude of SO_2 concentration at ground-level. Simulations at higher spatial resolution, relying on two nested domains with finest resolution of 7.3 km × 7.3 km, improve substantially the far-range vertical distribution of the volcanic cloud and subsequently the description of ground-level SO_2 concentrations. However, remaining discrepancies between model and observations are shown to result from an inaccurate representation of the planetary boundary layer (PBL) dynamics. Comparison with lidar observations points out a systematic under-estimation of the PBL height by the model, whichever the PBL parameterization scheme. Such a shortcoming impedes the capture of the overlying Bárðarbunga cloud into the PBL at the right time and in sufficient quantities. This study therefore demonstrates the key role played by the PBL dynamics in accurately modeling large-scale volcanogenic air pollution.

1 Introduction

On a local scale, the detrimental impact of volcanic gas, acid aerosol and ash emissions on the atmospheric environment (air pollution, rain acidification) and terrestrial ecosystems (soil, vegetation, groundwater, animals and humans) is well recognized (Delmelle, 2003; Hansell and Oppenheimer, 2004; Longo et al., 2008; van Manen, 2014; Horwell and Baxter, 2006; Ayris and Delmelle, 2012). However, volcanic sulphur-rich degassing can also generate air pollution events on a continental scale. Historical archives record evidences of long-range transport of acidic gases and aerosols from the 1783–84 Laki lava flood eruption (Iceland) up to Western and Central Europe (Thordarson and Self, 2003). Concomitantly, an abnormally high human mortality rate was observed not only in Iceland but also in Western Europe (Thordarson and Self, 2003; Witham and Oppenheimer, 2004; Grattan et al., 2005; Oppenheimer, 2011). In the specific case of the Laki eruption, it is difficult to draw a distinction between the respective impacts of volcanogenic air pollution and severe meteorological conditions, as extremes of heat and cold (which may have been partly caused by the eruption itself) occurred concurrently with the eruption (Oppenheimer, 2011). Nevertheless, there is little doubt that a Laki-style eruption would cause severe health hazards leading to an excess mortality rate at a continental scale (Schmidt et al., 2011). Obviously, at the time of the Laki eruption, only sparse observations on meteorological conditions (Yiou et al., 2014) and dispersed volcanic compounds (Thordarson and Self, 2003) were available, which hinders a thorough test of our ability to accurately model the dispersal of the prodigious emissions of volcanic SO_2 toward remote regions.

The long-lasting Holuhraun lava flood eruption (Aug 2014–Feb 2015) within the Bárðarbunga volcanic system (Iceland), hereafter called "Bárðarbunga eruption", allows for quantitatively assessing the far-range impact of a volcanic eruption on air quality. Even if of lesser magnitude than Laki (about one order of magnitude smaller in terms of emitted lava and sulphur degassing budgets (Gíslason et al., 2015)), the 6 month-long Bárðarbunga eruption continuously emitted abundant quantities of SO_2 into the lower troposphere reaching 11–12 Mt according to petrological estimates and ground-based UV-DOAS (Differential Optical Absorption Spectroscopy) observations (Gíslason et al., 2015). Whereas SO_2 air pollution is generally of anthropogenic origin, mainly associated with the combustion of sulfur-rich fossil fuels or with mining activities, the Bárðarbunga emissions have exceeded the budget of SO_2 emitted annually by all 28 state members of the European Union (4.6 Mt in 2011 (European Environment Agency, 2014)). Whereas SO_2 was released in large quantities, Bárðarbunga ash emissions were limited and therefore did not disturb air traffic, unlike the ash-rich 2010 Eyjafjallajökull and 2011 Grimsvótn icelandic eruptions. Nevertheless, Bárðarbunga volcano triggered an event of volcanogenic air pollution unprecedented in Europe in the modern era. Such pollution necessitated exceptional civil protection measures in Iceland (Gíslason et al., 2015). Indeed, high ground-level concentration of SO_2 and sulfate aerosols, mainly issued from the conversion of SO_2 in the atmosphere, is harmful to human health. SO_2 concentrations up to 9000–21000 $\mu g.m^{-3}$ were recorded in Iceland at a hundred kilometers from the eruption site, i.e. $\sim$60 times the hourly exposure limit value of 350 $\mu g.m^{-3}$ fixed by World Health Organization (WHO) (Gíslason et al., 2015).

The Bárðarbunga cloud travelled most often from the eruption site toward high latitudes, beyond the Arctic Polar Circle (Fig. 1 and supplementary material of McCoy and Hartmann (2015)). However, owing to favourable meteorological condi-

tions, the volcanic cloud was transported toward Western Europe in September 2014. This event fueled a far-range pollution event in SO_2 and particles which was recorded, without being exhaustive, in Fenno-Scandinavia (Ialongo et al., 2015; Grahn et al., 2015), Ireland, UK, the Netherlands (Schmidt et al., 2015) and France (Boichu, 2015). Contrary to stratospheric sulfate aerosols, few studies have allowed to fully determine microphysical properties of volcanic sulfates in aged tropospheric plumes (e.g. Bukowiecki et al. (2011) in the upper-tropospheric Eyjafjallajökull cloud in 2010), due to the difficulty to isolate the signature of sulfate from co-existing meteorological clouds and/or aerosols of a different nature. Here, we use a wealth of complementary observations from in-situ ground-level sampling (SO_2 and particulate matter), ground-based remote sensing (lidar, sun-photometry, UV-DOAS spectroscopy) available in Belgium and France, and satellite sensors (OMI and IASI) to characterize the distribution of volcanic SO_2 and sulfate aerosols as well as the dynamics of the planetary boundary layer (PBL). Both dynamical and microphysical properties of sulfate aerosols in the aged low-tropospheric Bárðarbunga cloud are provided.

We take advantage of this exceptional panel of observations to quantitatively examine and test our modeling ability to retrospectively reproduce the volcanogenic event of long-range air pollution taking place in late September 2014. While also relevant for industrial accident studies, such an exercise is critical to get prepared to accurately forecast a future large-scale episode of volcanogenic air pollution. Indeed, geological records indicate that Laki-style high-discharge lava flood eruptions, which emit huge amounts of sulphur compounds into the atmosphere, can occur in Iceland a few times per millennium (Thordarson and Larsen, 2007).

2 Methodology

2.1 Satellite/ground-based remote sensing and in-situ sampling

According to IASI observations of the altitude of Bárðarbunga SO_2 near Iceland (Fig. 1), the injection height is lower than 4-5 km. Consequently, the Level-2 product of the ultraviolet-visible OMI/Aura satellite sensor for the SO_2 total column (NASA GES DISC, 2016) is mostly preferred to hyperspectral infrared IASI/Metop observations whose sensitivity decreases below 5 km. In addition, the center of mass of the SO_2 cloud is assumed to be within the PBL. North-south gaps in snapshots of the SO_2 cloud result from the so-called 'row anomaly' of OMI detector (www.knmi.nl/omi/research/product/rowanomaly-background) which alters radiance data at all wavelengths for particular viewing directions. In a complementary manner, IASI captures on 21 September the front of the SO_2 cloud which is largely missed by OMI due to the 'row anomaly'. The major advantage of IASI is that it can track the altitude of SO_2 (Clarisse et al., 2014), even from moderate eruptions (Boichu et al., 2015).

[Figure 1 about here.]

A continuously-operating ground-based platform, with various remote sensing instruments, is installed on the roof of the Laboratoire d'Optique Atmosphérique in Lille-Villeneuve d'Ascq (northern France) and allows for tracking aerosols. It includes a micro-pulse CIMEL lidar measuring the radiation elastically backscattered by atmospheric particles and molecules at

532 nm. The BASIC algorithm (Mortier et al., 2013) allows for determining the vertical distribution of atmospheric particles over Lille as a function of time and distinguishing meteorological clouds from aerosols which are the focus of our study. A high load of low-tropospheric aerosols, lying at an altitude below 1.2 km, is highlighted and suspected to be partly or mostly of volcanic origin (Fig. 2).

Lidar observations are also used here to follow the PBL dynamics. The PBL is detected by applying a wavelet covariance transform to lidar backscatter profiles averaged over 20 minutes (Brooks, 2003). The PBL top is defined as the location of the maximum in the covariance profiles. As low-level meteorological clouds may disturb PBL height retrieval, a filter is applied so as to provide only cloud-free heights.

[Figure 2 about here.]

Complementary to lidar observations, the retrieval of ground-based sunphotometric observations, which are performed at two 80 km-distant sites (Lille and Dunkerque/Dunkirk), allows for identifying and isolating the signature of Bárðarbunga aerosols from other atmospheric particles transported over the north of France, such as cirrus particles here. Due to frequent cloudy conditions, time variations of vertically integrated aerosol properties derived from level-1.0 (not cloud-screened) and 2.0 (cloud-screened and quality assured) sunphotometric data from the AERONET network (Holben et al., 2001) are exploited using different inversion algorithms and a two-site approach (Lille and Dunkerque) (Fig. 3). Fine (sub-micron) and coarse (super-micron) aerosol optical depths (AOD) at 500 nm are retrieved using spectral deconvolution algorithm (SDA) applied on AOD within the range 340 to 1640 nm (O'Neill et al., 2003) (Fig. 3). A sharp and significant increase in the fine mode AOD is highlighted in the early afternoon of 21 September (Fig. 3), which will be shown later (Section 3.2) to correspond to the arrival of the Bardarbunga cloud over France. A persistent fine mode is then observed in the following days. Volume size distribution (VSD) of volcanic aerosols are determined using two different inversions: AERONET (version 2) standard algorithm which requires cloud-free almucantar observations (Dubovik and King, 2000; Dubovik et al., 2006) and recently developed GRASP (Generalized Retrieval of Aerosol and Surface Properties) code (Dubovik et al., 2014). Over the period of study, there is only one almucantar in Lille fulfilling AERONET level-2.0 requirements (on 23 September). Therefore, for the other two days, VSD is retrieved using GRASP: on 21 September, GRASP inverts a (manually inspected) cloud-free principal plane as in AERONET almucantar standard inversion (Torres et al., 2014); on 22 September, direct sun (DS) measurements (available without information of sky radiances) are inverted. For this latter inversion of DS observations, we assume VSD to be a bi-modal lognormal function and optical properties (i.e. refractive index and sphericity parameter) identical to those retrieved from the almucantar on 23 September. The consistency of these algorithms and strategies is shown in Fig. 4. Using a multi-site approach (ie., including AERONET VSD determined in neighbouring site of Dunkerque), the influence on the fine mode of cirrus co-existing with Bárðarbunga aerosols on 22 September in Lille is evidenced (Bottom of Fig. 4). SO_2 modeling in Fig. 8 shows that the volcanic cloud passes over Dunkerque (close to Calais) a few hours before Lille. Therefore, the similarity of fine-mode components retrieved at Lille and Dunkerque indicates that cirrus in Lille weakly influence the fine-mode, which is in turn mainly associated with volcanic sulfate aerosols in this specific case.

[Figure 3 about here.]

[Figure 4 about here.]

Daily ground-based MAX(Multi-AXis)-DOAS observations in the ultraviolet are performed by BIRA-IASB in Brussels-Uccle (Belgium) and provide time series of SO_2 column amounts during daylight hours (time step of $\sim$ 12 min). The instrument is described in Gielen et al. (2014) while retrieval method and settings can be found in Wang et al. (2014).

Ground-level concentrations of SO_2 and particles are routinely measured in France by a network of ground stations managed by accredited associations responsible for air quality monitoring. For this study, AIRPARIF provided observations at Neuilly-sur-Seine (near Paris) and Atmo Nord–Pas-de-Calais at Calais and Lille-Fives (northern France). Ground-level SO_2 concentrations are monitored by ultraviolet fluorescence with a time step of 15 min. Mass concentration of particulate matter, with diameters less than 2.5 μm (PM2.5) and 10 μm (PM10), are measured by TEOM-FDMS (Tapered Element Oscillating Microbalances with Filter Dynamics Measurement Systems) (time step of 15 min) or by RST (Regulated Sampling Tube) beta gauge automated air monitors (time step of 2 hours) which account for both volatile and non-volatile PM fractions.

2.2 Meteorological and chemistry-transport models

The atmospheric dispersion of volcanic SO_2 is described using the CHIMERE Eulerian regional chemistry-transport model (CTM) (Boichu et al., 2013, 2014, 2015). The model accounts for various physico-chemical processes affecting the SO_2 released in the atmosphere, including transport, turbulent mixing, diffusion, dry deposition, wet scavenging and gas-/aqueous-phase chemistry. However, the conversion of SO_2 to sulfate aerosols is not implemented in this study to avoid uncontrolled influence of uncertainties on the numerous factors governing this process in a volcanic cloud as they have not been specifically validated for application to a volcanic plume. Not accounting for this process leads to underestimate the actual volcanic emissions. CHIMERE CTM is driven by meteorological fields from the Weather Research and Forecasting (WRF) model (Skamarock et al., 2008), which is forced by NCEP (National Centers for Environmental Prediction) reanalysis data on a 6-h basis (Kalnay et al., 1996). The spin time up of WRF simulations is of five days. WRF meteorological fields have a 25 km $\times$ 25 km horizontal grid and 30 hybrid sigma-pressure vertical layers extending up to $\sim$ 19 km above sea level (a.s.l.). The dynamics of the PBL is described by the Yonsei University (YSU) parameterization scheme, which is the most widely used scheme implemented in WRF (Hong et al., 2006). It consists of a first-order, non-local scheme with an explicit entrainment layer and a parabolic K-profile in an unstable mixed layer. The calculated PBL height is then used as an input to CHIMERE. CHIMERE simulations are performed over the period 19–24 September 2014 on a large area extending from North of Greenland down to Spain. CHIMERE CTM has the same horizontal resolution as WRF but a finer vertical resolution with 29 hybrid sigma-pressure vertical layers extending up to 150 hPa ($\sim$ 13 km a.s.l.).

SO_2 emissions are poorly known. For simplicity, we model the source term as a step-function in time with an amplitude of 4700 t.h^{-1}, which roughly corresponds to peak values of the SO_2 flux retrieved from ground-based UV-DOAS spectroscopy (Gíslason et al., 2015). SO_2 is released along a Gaussian profile with a full width at half maximum of 100 m. Time of release

and altitude of emissions are found by trial and error so as to reproduce by visual inspection first-order features of satellite and ground-level SO_2 observations. As represented in Fig. 5, we find that two step-functions at (1) 1 km a.s.l. from 19 September 2014 12:00 UT until 24 September 2014 00:00 UT and (2) at 4 km a.s.l. from 20 September 2014 12:00 UT until 24 September 2014 00:00 UT, are sufficient to fit the two-wave behaviour of the Bárðarbunga cloud (Fig. 6). This upper injection height is consistent with IASI level 2 products of SO_2 altitude, which captured the Bárðarbunga SO_2 cloud in the vicinity of the source on 19, 20, 22 and 23 September 2014 (Fig. 1). Accordingly, this source term is not intended to reflect the full complexity of the actual emissions of Bárðarbunga but rather captures only the SO_2 parcels traveling toward Western Europe.

[Figure 5 about here.]

3 Results

3.1 Large-scale dispersal of SO_2 toward Europe

According to OMI satellite observations, the model reproduces the correct timing of SO_2 arrival in northern Scotland on 20 September 2014 descending down to the south-western coast of England on 21 September (Fig. 6). SO_2 observed to the north of 60°N and to the east of 5°W on 21 September is absent from simulations because these emissions, which were released at a high altitude (above 8 km a.s.l according to IASI images of the SO_2 altitude in Fig. 1), are not accounted for in the model source term as they are first transported toward Arctic then dispersed toward Fenno-Scandinavia, i.e. out of our domain of interest (i.e Western Europe). The model indicates a first SO_2 wave (wave 1 in Fig. 6 and movie of the modeled dispersal of the Bárðarbunga SO_2 cloud in Supplementary Material) reaching Belgium and northern France on 21 September, which cannot be confirmed by OMI observations, hampered above France due to north-south gaps resulting from detector 'row-anomaly'. It is however captured by IASI (inset in Top of Fig. 6), which also indicates that this wave travels at a low altitude below 5 km a.s.l. (Fig. 1). This first wave is then pushed and dispersed toward the Atlantic Ocean on 22 September. Note that some traces of SO_2 detected by OMI over mainland Europe on 22 September are not associated with wave 1 but likely result from emissions released and then transported at high altitudes (above 8 km a.s.l.) toward Fenno-Scandinavia according to IASI images of the volcanic SO_2 altitude (Fig. 1). However, these emissions, as stated earlier, are not accounted for in the model. As a result, these traces of SO_2 over mainland Europe cannot be reproduced by simulations. On 22 September, both model and observations depict a north-south elongated SO_2 cloud reaching France for the second time (wave 2 in Fig. 6 and movie in Supplementary Material). Modeled SO_2 column amounts are in agreement with OMI SO_2 loading. However, the absence of SO_2 above Fenno-Scandinavia in the OMI image contradicts the model. This inconsistency may result from inaccuracies of prescribed altitudes of SO_2 injection or of meteorological forcing of the model. On 23 September, the model shows the arrival of SO_2 above Norway/Sweden, after a long transport from Iceland up to northern Greenland. On the same day, model and observations both indicate some dispersed remnants (although of different intensity) of the second SO_2 wave having hit western Europe above western France and southern UK.

[Figure 6 about here.]

3.2 Arrival of the volcanic cloud in the far-range lower troposphere

The precise timing of arrival of the Bárðarbunga cloud in the French lower troposphere on 21 September 2014 is deduced from the synergetic analysis of volcanic SO_2 modeling as well as observations from ground-based lidar and sunphotometers which remotely sense aerosols, on a continuous basis, above Lille-Villeneuve d'Ascq. Sunphotometry indicates the arrival of fine mode aerosols between 12:00 and 15:30 UT on 21 September (Fig. 7-a1), presumably sulfate aerosols formed from volcanic SO_2 in the atmosphere, according to their high single scattering albedo (~ 0.98) derived from AERONET inversions indicative of non- or weakly-absorbing aerosols. While fine mode AOD values remain below 0.1 at midday in Lille on 21 September, the arrival of volcanic sulfate aerosols marks an increase in AOD with values ranging between 0.3 and 0.45 in the afternoon (Fig. 3). Principal plane inversion also provides volume size distribution (Fig. Fig. 7-a2), with an effective radius (r_{eff}) of these sulfate of 0.21 μm (mean volume radius r_v of 0.26 μm). Simultaneously, lidar active observations, which characterize the temporal evolution of aerosol vertical distribution, indicate the presence above Lille of aerosols at 2 km a.s.l., with a decreasing altitude with time (Fig. 7-b). This behavior of aerosols coincides with the temporal decrease of the modeled altitude of the most concentrated layer of volcanic SO_2 accompanying the first SO_2 wave described in Section 3.1 (red line in Fig. 7-b). This common evolution evidences the co-existence of SO_2 and sulfate aerosols within the low-altitude volcanic cloud. Soon thereafter, ground-level sampling in Lille records the first significant increase of SO_2 concentration up to ~ 20 μg.m^{-3} (compared to background values usually close to zero at this site except when contaminated by nearby urban heating plant) followed by a first rise in particulate matter abundance up to ~ 35 μg.m^{-3} (Fig. 7-c). Hence, these four pieces of evidence (SO_2 modeling, sunphotometry, lidar and ground-level air sampling) unambiguously confirm the arrival of the Bárðarbunga cloud in the French lower troposphere down to the ground in the early afternoon of 21 September.

After a period of quiescence, a second, more prolonged and intense episode of ground-level air pollution, in both SO_2 and particles, is recorded from 22 to 23 September in Lille (Fig. 7-c). During this second episode, the PM concentration exceeds the information and recommendation threshold prescribed by WHO of ~ 50 μg.m^{-3}, defined as the hourly running 24 hour average value. Concomitantly, sunphotometry indicates a persistent fine-mode (Fig. 7-a2) of weakly absorbing aerosols, which produce fine mode AOD values abnormally high for Lille and Dunkerque (up to ~ 0.8, Fig. 3). The size of Bárðarbunga sulfate aerosols (r_{eff} within 0.26–0.28 μm, r_v within 0.21–0.24 μm) largely exceeds the radius characterizing typical urban aerosols in Lille ($r_{eff} < 0.2$ μm (Mortier, 2013)). This size is also larger than values reported by sparse observations of volcanic tropospheric sulfate radius at distance from the volcanic source (r_v within 0.12–0.16 μm in the Eyjafjallajökull cloud (Bukowiecki et al., 2011)).

[Figure 7 about here.]

3.3 Far-range air pollution at ground level

Substantial increases in ground-level SO_2 concentration are recorded by air quality monitoring networks not only in the north end of the country but also on a broad regional scale in France. Unseen for more than a decade, this makes this event of SO_2 pollution exceptional (Fig. 8). Interestingly, this pollution episode strictly follows a similar temporal pattern, except for a

time lag, whichever the city of observation. As shown from the combined analysis of space-based SO_2 observations and CTM simulations at a large scale (Section 3.1 and movie of the modeled dispersal of the Bárðarbunga SO_2 cloud in Supplementary Material), this temporal behaviour results from the arrival of two successive waves of SO_2 reaching France from 21 to 23 September. At ground-level, air quality measurements track the progressive transport of these two waves from the north to the center of France (blue lines in Fig. 8). SO_2 concentrations up to 70 μg.m^{-3} are associated with the second wave, which is recorded firstly in Calais, then successively 3 hours later in Lille-Fives and 8 hours later much further south near Paris in Neuilly-sur-Seine. While the modelled time series of SO_2 column amounts reproduce this two-wave pattern (solid red line in Fig. 8), simulations fail in correctly describing ground-level SO_2 concentration as the second wave of pollution starting on 22 September is missed (dashed red line in Fig. 8).

[Figure 8 about here.]

4 How to improve long-distance air quality modeling?

4.1 Limitations of simulations with a standard configuration

As illustrated by the broad agreement with OMI satellite data (Fig. 6 and Section 3.1), chemistry-transport simulations, with a standard configuration here, are efficient at reproducing on a continental scale the dispersion of the Bárðarbunga SO_2 cloud from Iceland toward France. The temporal dynamics of far-range events of air pollution, characterized for instance in France by the arrival of two distinct SO_2 waves traveling from north toward the capital city in $\sim$ 8 hours, is hence well described (solid red line in Fig. 8). A good agreement is also reached between model and observations of SO_2 vertical column amounts (CA) by ground-based UV max-DOAS spectroscopy performed at long distance from the eruptive site in Uccle/Belgium, located less than 100 km from Lille/France (Fig. 9). Model and observations find SO_2 CA of the same magnitude on 19, 20, 22 and 23 September. Nevertheless, the model cannot capture the significant and abrupt SO_2 CA increase (up to 14 DU) of very short duration (from 14:18 to 15:07 UT) observed on 21 September by DOAS, likely due to the insufficient (hourly) time resolution of the model.

[Figure 9 about here.]

However, the model has difficulty reproducing the correct intensity of the air pollution episode in remote areas. Similar issues have also arisen with independent modeling simulations using a Lagrangian approach forced with distinct meteorological reanalysis(Schmidt et al., 2015). Our model here completely misses the second wave of SO_2 at ground-level in the north of France (dashed red line in Fig. 8). This shortcoming results from an incorrect description of the vertical distribution of SO_2 at long distance from the eruptive site. According to lidar observations capable to detect sulfate aerosols coexisting with SO_2 (Section 3.2), the model mimics correctly the drop in altitude above Lille of the first SO_2 wave on 21 September (red line in Fig. 7-b). This modeled wave hits the surface at about the same time as the first detection of air pollution at ground-level. But the second modeled wave, despite a similar pattern with a significant decrease in altitude with time from 6 km a.s.l., does not reach the ground and remains at an altitude $\geq$ 1.8 km above Lille on 22 September (red line in Fig. 7-b).

Issues encountered for adequately modeling far-range air pollution episodes can arise from the difficulty of simulating both the long-range transport/dispersal of volcanic compounds and the meteorological dynamics at a local scale, as the latter controls the capture and mixing of the overlying volcanic cloud in the far-range planetary boundary layer (PBL). Simulations at higher spatial resolution of both CHIMERE CTM and WRF models may help to make progress along this path. For these reasons, we explore in the next sections the impact on far-range ground-level SO_2 concentrations of both meteorological/chemistry-transport simulations at higher spatial resolution and of various PBL parameterization schemes in the meteorological model.

4.2 Improvements reached with simulations at higher spatial resolution

Meteorological and chemistry-transport simulations at higher spatial resolution require both high computation time and capacity, which challenges our current modeling capacities. We performed here WRF and CHIMERE simulations on two nested horizontal grids (Fig. 10). The larger domain extends from north of Greenland down to Spain (as the low resolution domain of simulations in Sections 2.2 and 3) with a horizontal resolution of 22 km $\times$ 22 km representing 209 $\times$ 229 grid cells. Note that this coarse resolution is nevertheless slightly higher than the low spatial resolution simulation performed on a 25 km $\times$ 25 km horizontal grid. The nested domain extends from Norway down to Central France, with a fine 7.3 km $\times$ 7.3 km horizontal resolution representing 217 $\times$ 232 grid cells in a Lambert projection. Except for one test run configured with 60 hybrid sigma-pressure vertical layers extending up to $\sim$ 19 km a.s.l. and $\sim$ 13 km for WRF and CHIMERE models respectively, most simulations are run assuming 30 vertical layers for both models, as in the simulations with a standard configuration in Section 3. The dynamics of the PBL is still described by the Yonsei University (YSU) scheme, as in Section 3.

[Figure 10 about here.]

Simulations at higher horizontal spatial resolution better resolve the long-distance transport of the Bárðarbunga SO_2 cloud as well as its descent over France. Especially, an earlier and faster descent of the SO_2 cloud over Lille is modeled on 22 September 2014, with the core of the plume reaching a significantly lower altitude than in low resolution simulations (Top panels of Fig. 11). Subsequently, a clear improvement of modeled far-range ground-level SO_2 concentrations is reached. Indeed, this earlier modeled descent of the SO_2 cloud leads to the emergence of a second peak in ground-level concentrations on 22 September (Bottom panel of Fig. 11-right), which was entirely missed by simulations at low spatial resolution as mentioned in Section 4.1 (Bottom panel of Fig. 11-left).

[Figure 11 about here.]

The emergence of a second peak is also modeled at other air quality monitoring stations, i.e. Calais and Neuilly-Sur-Seine (Fig. 12). A better agreement between model and observations is also noticed regarding the timing of the first peak concentration whichever the station (Fig. 12). Note that only slight differences in ground-level concentrations were observed with simulations performed with a twice higher vertical resolution (i.e. 60 vertical layers in WRF and CHIMERE models) and are consequently not shown.

[Figure 12 about here.]

Nevertheless, although clear improvements in ground-level SO_2 concentrations are achieved with simulations at higher spatial resolution, both timing and intensity of the second peak concentration are not perfectly reproduced by the model (Fig. 12). The emergence of the second peak concentration is modeled late compared to measurements at Calais and Lille, and slightly too early at Neuilly-sur-Seine. The modeled intensity of this peak concentration is always under-estimated by a factor of 3 to 10 depending on the monitoring station.

These remaining discrepancies between model and observations may arise from various reasons. We explore in the next sections the impact on far-range ground-level concentrations of (i) a poor knowledge of the source emissions (assumed here to follow a simple pattern as described in Section 2.2), and (ii) an incorrect modeling of the planetary boundary layer dynamics which may prevent from correctly capturing the descending volcanic cloud down to the ground.

4.3 Minor role of source term variations

We do not aim to provide a detailed estimate of the source term but to show that a simple source term allows for representing the main features of this event of far-range air pollution triggered by Bárðarbunga eruption (Sections 3.1 and 3.3). Instead, we investigate here whether variations in this simple source term could explain the current discrepancies between observed and modeled far-range SO_2 concentrations at ground-level.

Whichever the monitoring station, we have deduced from SO_2 modelling that the second peak in ground-level SO_2 concentration results from the arrival in France of the second pulse of emissions, which are injected at the source at 4 km a.s.l. from 20 September 2014 12:00 UT until 24 September 2014 00:00 UT in low spatial resolution simulations (not shown). We vary the altitude of injection (between 3 and 7 km a.s.l.) as well as the start time of this second pulse of emissions (with a 5-hour earlier release), as both may impact the timing of the second peak concentration which is not correctly reproduced by the model (Section 4.2 and Fig.12). However, only slight modifications on far-range ground-level concentrations are obtained, unable to explain the discrepancy observed between model and observations.

Regarding the under-estimation by the model of ground-level SO_2 concentrations by a factor of 3 to 10 (Section 4.2), one could argue that it results from an under-estimation of the assumed SO_2 emission flux by a similar factor given linear processes of large-scale transport/dispersion. Volcanic emissions may present rapid temporal fluctuations of large amplitude (e.g Boichu et al. (2013)). However, despite a poor time-resolved knowledge of the Bárðarbunga SO_2 source relying on sparse ground-based measurements, assuming a SO_2 flux five time stronger would better fit far-range ground-level concentrations but would also lead to far-range SO_2 column amounts increased by the same amount (not shown). The latter would be in complete disagreement with SO_2 column amounts retrieved from satellite observations (Fig. 6) or ground-based MaxDOAS measurements performed in Belgium (Fig. 9).

As a consequence, input model parameters characterizing the Bárðarbunga SO_2 source (flux and altitude of injection) are shown to play a minor role on far-range ground-level concentrations over our relatively short period of study (19-24 September 2014). They do not allow us to solve the disagreement observed between model and observations. For this reason, we explore in the next section the impact of the PBL dynamics on air quality modeling.

4.4 Key role of the planetary boundary layer

In addition to the reference Yonsei University (YSU) scheme used in the low spatial resolution simulation, the impact on far-range ground-level concentrations of two additional PBL parameterization schemes, recently added to the WRF model, are tested: the Asymmetric Convective Model (ACM2) scheme (Pleim, 2007) as well as the improved Mellor-Yamada-Nakanishi-Niino level 3 model (MYNN 3) scheme (Nakanishi and Niino, 2006). The ACM2 scheme is a first-order, non-local closure scheme which features non-local upward mixing and local downward mixing. The MYNN 3 scheme is a second order, local closure scheme tuned to a database of large-eddy simulations.

At first glance, time series of the PBL height above Lille do not seem to vary widely with the different PBL parameterization schemes (Top of Fig. 13). We note nevertheless a less marked diurnal cycle with the MYNN3 scheme. However, these slight differences are shown to be sufficient to produce up to a ten-fold variation of the ground-level SO_2 concentrations (Bottom of Fig. 13).

[Figure 13 about here.]

As illustrated by Fig. 14, both timing and altitude of the encounter of the boundary layer top and the overlying volcanic SO_2 cloud, which may vary with the PBL parameterization scheme, strongly impact the subsequent increase in SO_2 concentration at ground-level some time later.

For our specific case-study, the top of the PBL encounters the overlying Bárðarbunga SO_2 cloud above Lille at approximately the same time on 21 and 22 September, whichever the PBL scheme (Fig. 14). After this capture of the volcanic cloud into the PBL, SO_2 is mixed and diffused down to the ground triggering a noticeable increase of the ground-level SO_2 concentration. The time delay between the capture of the volcanic SO_2 at the top of the PBL and its record at the ground-level is estimated of just a few hours (Fig. 14).

However, only the ACM2 scheme allows the top of the PBL to encounter the core (i.e. the most concentrated part) of the Bárðarbunga SO_2 cloud on 22 September 2014, with a PBL height at the time of encounter higher by just a few hundred of meters compared to other schemes (Top right of Fig. 14)). This substantial capture of volcanic SO_2 by the boundary layer explains why the intensity of the second SO_2 peak concentration is the highest with this scheme and the closest to observations (Bottom right of Fig. 14). Note that the best agreement between observations and model for the first SO_2 peak concentration is also reached with the ACM2 scheme. These results demonstrate the crucial importance to correctly model both the PBL

height and the vertical distribution of the overlying volcanic SO_2 cloud with time. Note that this latter depends on a rigorous modeling of both long-distance transport/dispersion processes and of local PBL dynamics. Indeed, the PBL scheme influences the concentration of the overlying volcanic SO_2 (Top of Fig. Fig. 14). We may even suspect a kind of "sucking" of the core of the volcanic cloud which seems to follow the PBL top, especially remarkable on 22 September.

[Figure 14 about here.]

Nevertheless, even if the ACM2 scheme provides the best fit to observations, none of the PBL schemes allows for precisely modeling the second SO_2 peak concentration with correct timing and intensity (Fig. 13). This difficulty likely results from the inaccuracy of the modeled PBL height which presents marked differences with observations, whichever the PBL scheme. The altitude of the PBL above Lille is retrieved from lidar observations and compared with model output in the bottom of Fig. 15. Simulations generally underestimate the PBL height over Lille (up to 1.5 km), especially in the mornings and evenings. Such underestimation is a relatively common feature of WRF PBL schemes (Banks et al., 2015), used here to force the CHIMERE chemistry-transport model.

In a context of urban air pollution where pollutants are injected at ground-level into the atmosphere, an underestimation of the PBL height favors an over-evaluation of the intensity of ground-level pollution. In our volcanic case-study, this may explain the overestimation by certain schemes of the SO_2 ground concentration increase resulting from the first SO_2 wave reaching the ground of Lille on 21 September in the evening (Fig. 13). However, the PBL height underestimation by the model can also prevent from correctly capturing in the PBL the second SO_2 wave which travels at a higher altitude than the first wave (Fig. 14). In this context, the intensity of air pollution at ground-level is under-evaluated (Bottom of Fig. 13).

In our specific case-study, our concern is that, whichever the scheme, the modeled PBL height increases too lately and too weakly compared to lidar observations in Lille, which is especially problematic in the morning of 22 September (Bottom of Fig. 15). Indeed, this discrepancy explains both the delayed modeled timing of the second peak concentration and a substantial under-estimation of its intensity (by a factor of 2-3), as the modeled boundary layer captures too late a smaller fraction of the overlying volcanic SO_2 than it should in reality. In other words, an earlier and higher modeled PBL height in the morning of 22 September, as expected according to lidar observations, would lead to an earlier and stronger capture of the overlying Bárðarbunga SO_2 cloud at the top of the boundary layer. This would produce an earlier and stronger peak concentration at ground-level in better agreement with air quality monitoring observations.

Therefore, this case-study demonstrates the key role played by the PBL dynamics to rigorously estimate the magnitude of far-range volcanogenic air pollution.

[Figure 15 about here.]

5 Conclusions

The Bárðarbunga eruption provides the exceptional opportunity to carry out a modelling exercise of a far-range volcanogenic air pollution event using a broad panel of complementary measurements acquired by space and ground-based (remote sensing and in-situ) sensors.

Chemistry-transport modeling reproduces the large-scale dispersal of SO_2 from Iceland toward western Europe as observed from satellite OMI and IASI sensors. The synergetic analysis of SO_2 modeling and aerosol dynamics deduced from sunphotometric and lidar observations allows us to determine the exact timing of arrival of the volcanic cloud in the distant lower troposphere of northern France before its descent to the ground. The joint analysis of lidar measurements with the retrieval of multi-site sunphotometric observations using recently-developed inversion algorithms also provides a full characterization of volcanic sulfate aerosol properties with time (loading, vertical repartition, size distribution and single scattering albedo).

Based on this combined analysis of volcanic SO_2 and sulfate aerosols, we highlight the success and the challenges in simulating far-range episodes of air pollution. We show that the air pollution triggered by the Bárðarbunga eruption in late September 2014 is characterized by the arrival to France of two distinct SO_2 waves. The descent of these waves down to the ground produces two substantial peak concentration recorded at different monitoring ground stations in France with a time lag of 3 to 8 hours. The specific temporal pattern of this pollution event is well described even with low (25 km $\times$ 25 km) horizontal spatial resolution simulations. However, the model faces difficulties in reproducing the correct magnitude of one of the two ground-level SO_2 peak concentrations.

We show that large improvements on the far-range vertical distribution of the dispersed volcanic cloud and subsequently on surface concentrations are gained with simulations carried out at higher spatial resolution. Such simulations rely on two nested horizontal grids, which include a large domain with a coarse resolution of 22 km $\times$ 22 km and a narrower domain with a fine resolution of 7.3 km $\times$ 7.3 km. High computational capacities are required given the very large extent of the area flown over by the Bárðarbunga volcanic cloud in late September 2014, from northern Greenland down to south of France. Nevertheless, some discrepancies remain as high spatial resolution simulations do not reproduce correctly the timing of the second SO_2 peak concentration at ground-level (with a difference of a few hours) and the intensity of this peak is substantially under-estimated compared to observations.

The reasons for these remaining discrepancies between model and observations of far-range ground-level concentrations are investigated. Variations in the source term parameters (i.e. flux and altitude of injection) are shown to have a minor impact during the period of time of our study. However, the PBL dynamics plays a key role. Testing three parameterization schemes for the planetary boundary layer in the WRF model (YSU, ACM2 and MYNN3), a resulting ten-fold variability of surface concentrations is obtained. The ACM2 scheme provides the best fit to observations. Nevertheless, it does not perfectly repro-

duce the timing and intensity (under-estimated by a factor 3) of the second peak concentration. Lidar observations performed in Lille allows us to test the validity of the modeled PBL height time series at this location. During the morning of specific interest, the modeled PBL height increases too late and too weakly compared to observations. This shortcoming results in a too late and too weak capture of the overlying Bárðarbunga SO_2 cloud by the boundary layer and, subsequently, a delayed peak concentration at ground-level with an under-estimated intensity.

This case-study points out how fundamental it is to simulate accurately the PBL dynamics for modeling large-scale volcanogenic air pollution. Such difficulties will need to be overcome in order to get prepared to accurately forecast far-range air pollution episodes triggered by future eruptions releasing large amounts of toxic gases to the atmosphere.

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List of Figures

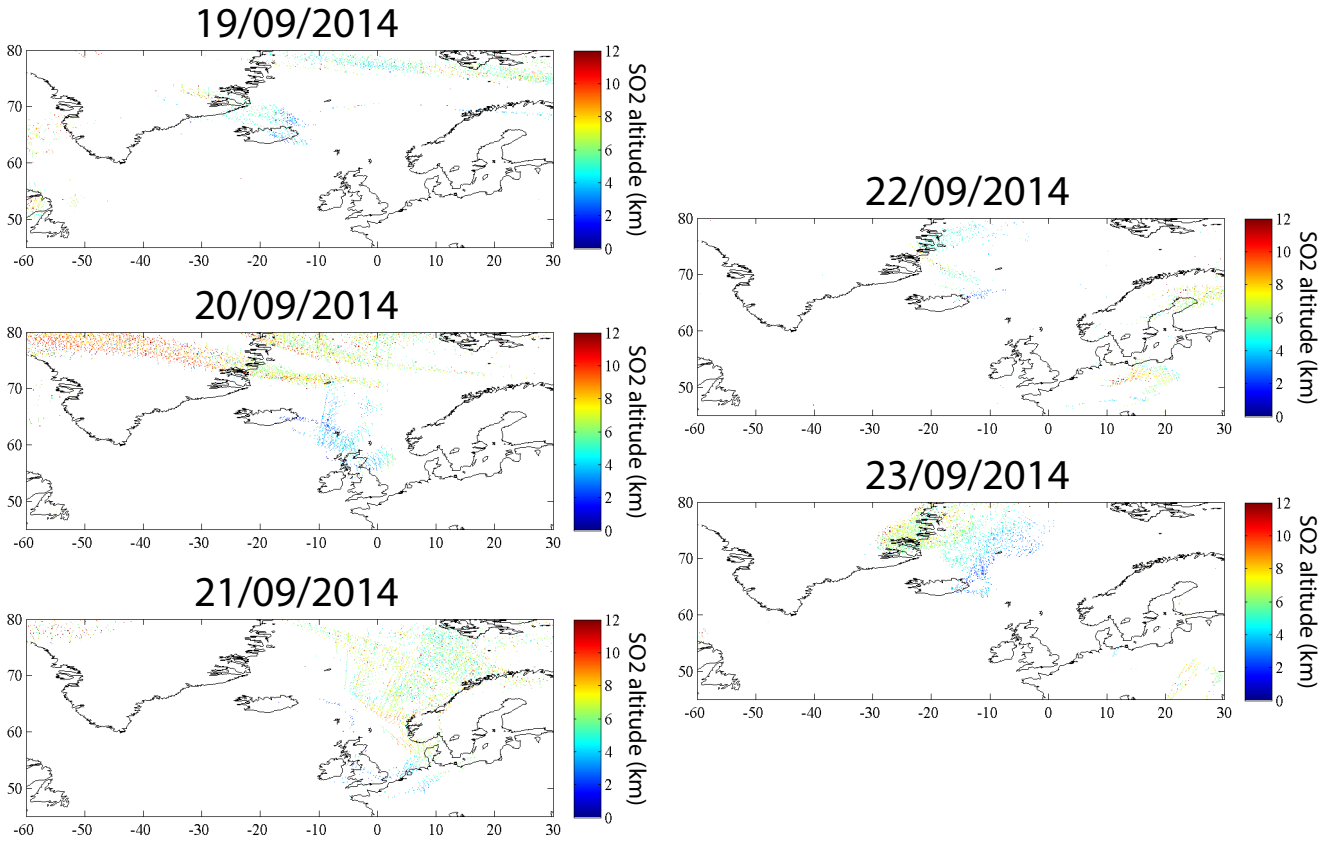

Figure 1. Altitude (in km a.s.l.) of Bárðarbunga SO_2 retrieved from IASI observations.

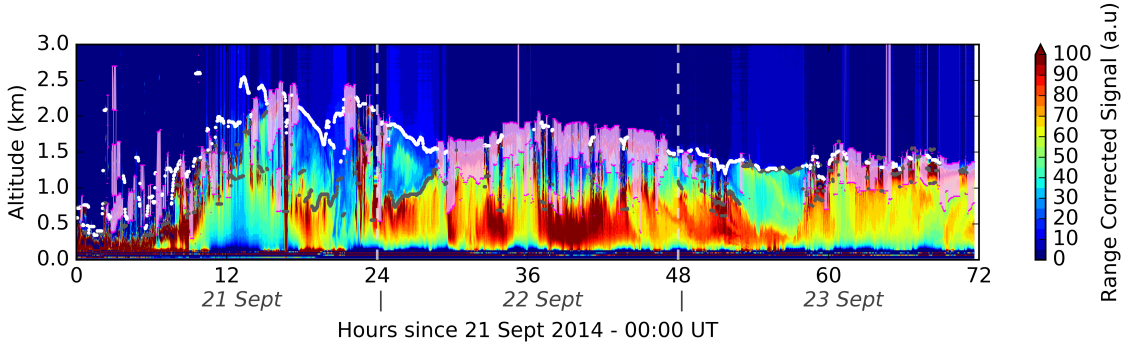

Figure 2. Vertical distribution of atmospheric particles detected over Lille by lidar observations performed from 21 to 23 September 2014. The BASIC algorithm detects meteorological clouds above 300 m a.s.l. (pink), the top of the aerosol layer (white) and a heavy load of low-tropospheric aerosols including volcanic particles (lying at an altitude below ∼1.2 km), which is characterized by a strong backscatter signal (yellow/red).

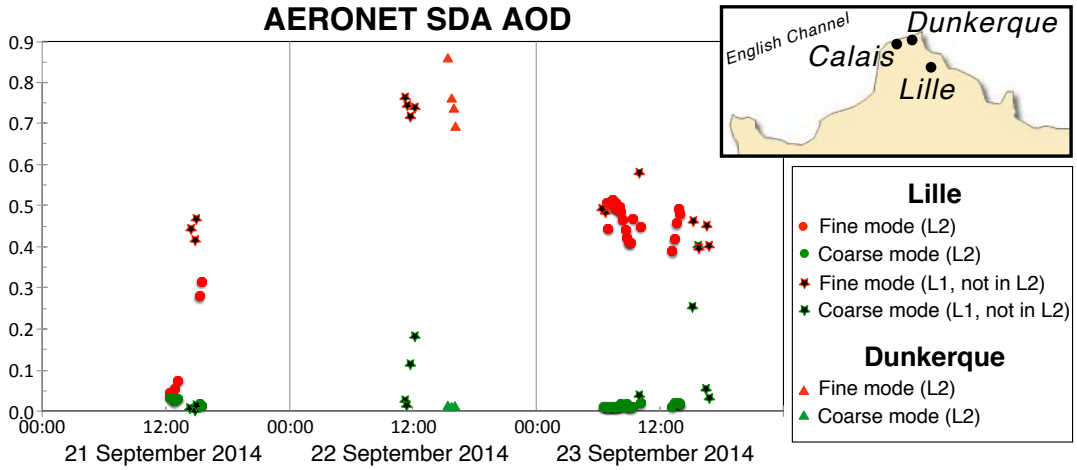

Figure 3. AERONET sunphotometric fine and coarse mode aerosol optical depth (AOD) at 500 nm retrieved using SDA algorithm in Lille and Dunkerque from 21 September to 23 September 2014. Map of northern France in inset.

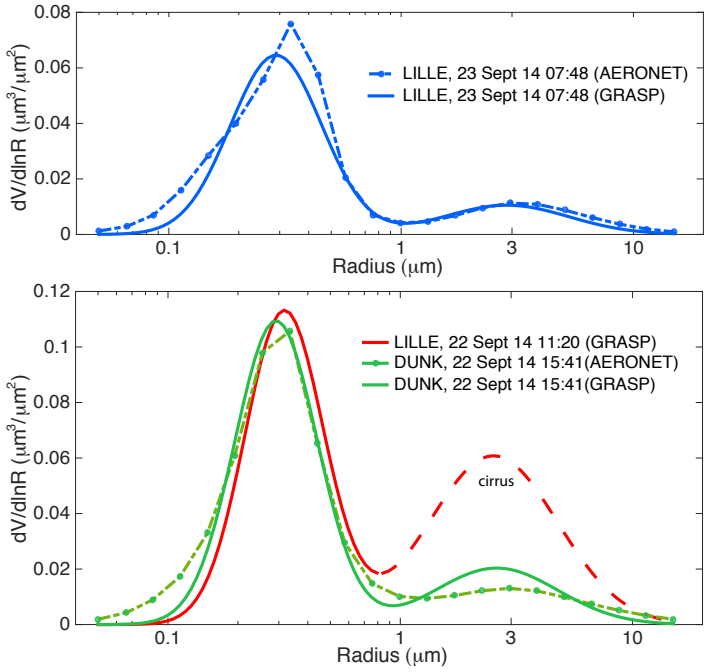

Figure 4. (Top) Consistency of the volume size distribution (VSD) in Lille on 23 September retrieved (dashed line) by inversion of Almucantar observations using standard AERONET inversion and (solid line) by inversion of direct sun measurements using GRASP algorithm. (Bottom) For 22 September, consistency of VSD retrieved by inversion of almucantar using standard AERONET inversion in Dunkerque (green dashed line) and by inversion of direct sun measurements using GRASP algorithm in Dunkerque (green plain line) and Lille (red line).

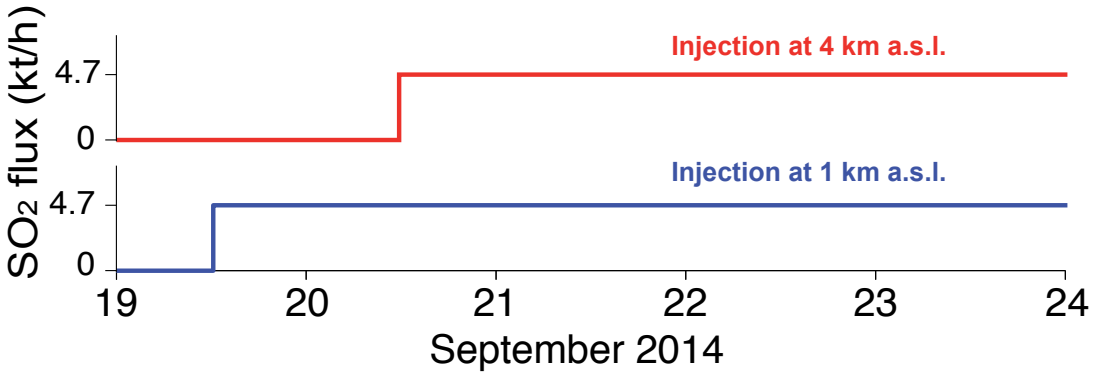

Figure 5. Simple source term (i.e. flux and altitude of injection of emissions as a function of time) initializing chemistry-transport simulations of the Bárðarbunga SO_2 cloud dispersal toward Europe in late September 2014.

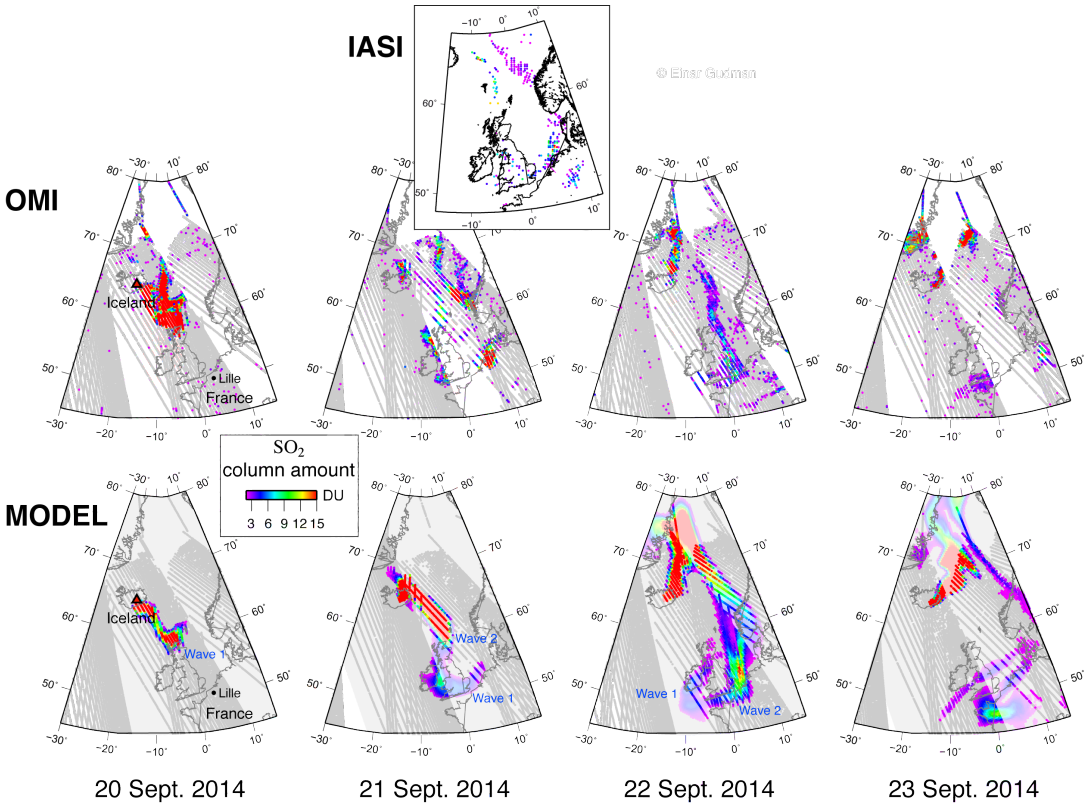

Figure 6. Dispersion of SO_2 from Bárðarbunga eruption toward Europe in late September 2014 (top) observed from satellite imagery (timeseries of OMI PBL products and IASI data in inset for 21 September) and (bottom) modeled using CHIMERE chemistry-transport model. Grey points indicate OMI column amounts < 2 DU. White zones show areas where data are not available. To facilitate the comparison between model and observations, the model is displayed transparently over zones where OMI data are not available.

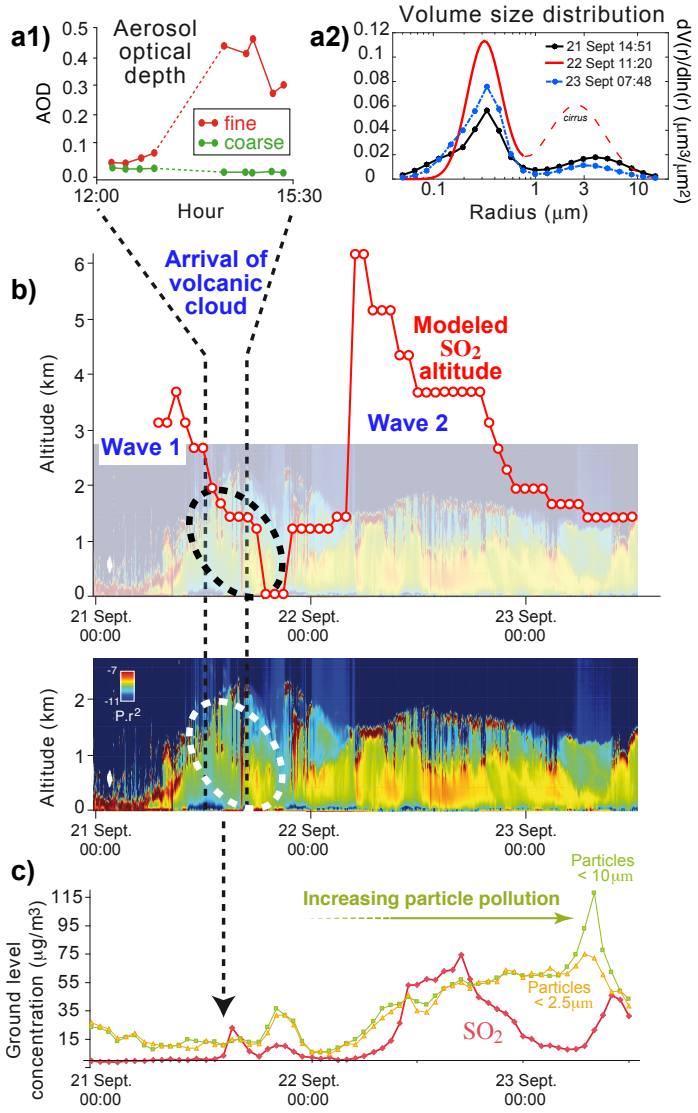

Figure 7. Multi-parametric observations and modeling of SO_2 and aerosols in Lille: (a) Retrieval of sunphotometric observations yields (a1) aerosol optical depth (at 500 nm) for coarse (green) and fine (red) mode on 21 September and (a2) time variations of aerosol volume size distribution. (b) Time series of the modeled altitude (red line) of the most concentrated layer of volcanic SO_2 overlaid on the lidar range-corrected backscatter signal ($\ln(P.r^2)$) at 532 nm. Distinction between aerosol and meteorological clouds in lidar data is shown in Fig. 2. (c) Ground-level concentration of SO_2 (red) and particles (PM2.5 in yellow and PM10 in green).

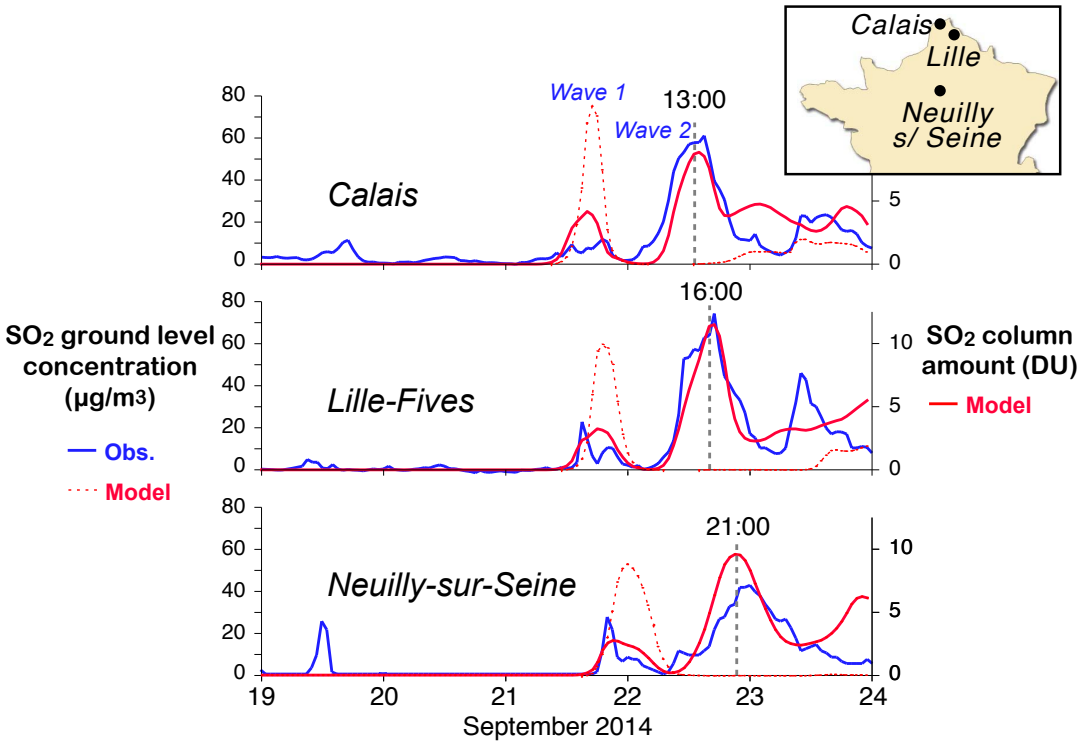

Figure 8. SO_2 ground-level concentration observed by air quality networks in France (blue) and modelled (dotted red), compared with modelled SO_2 vertical column amount (solid red).

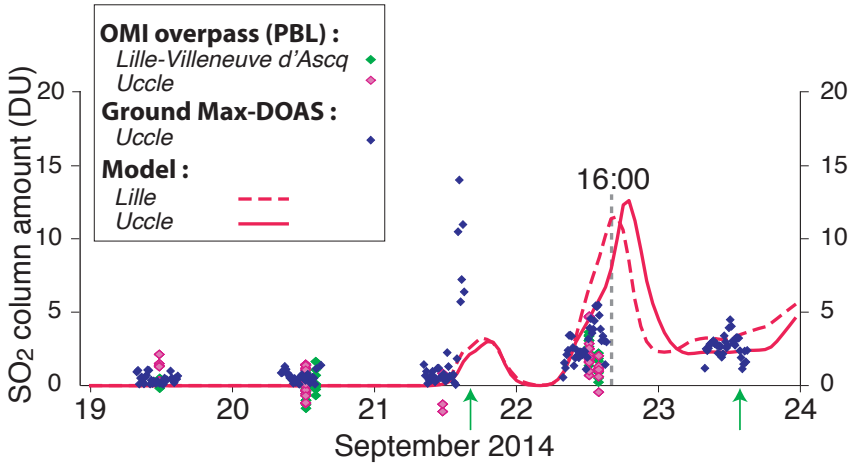

Figure 9. Time series of SO_2 vertical column amount above Uccle (Belgium) and Lille (France) from CHIMERE CT model (dashed and solid red lines for Lille and Uccle resp.), OMI PBL overpass (green and pink diamonds for Lille and Uccle resp.) and ground-based UV MAX-DOAS observations in Uccle (blue diamonds). Green arrows indicate when OMI overpasses above Lille are missing due to gaps in data related to sensor 'row anomaly'.

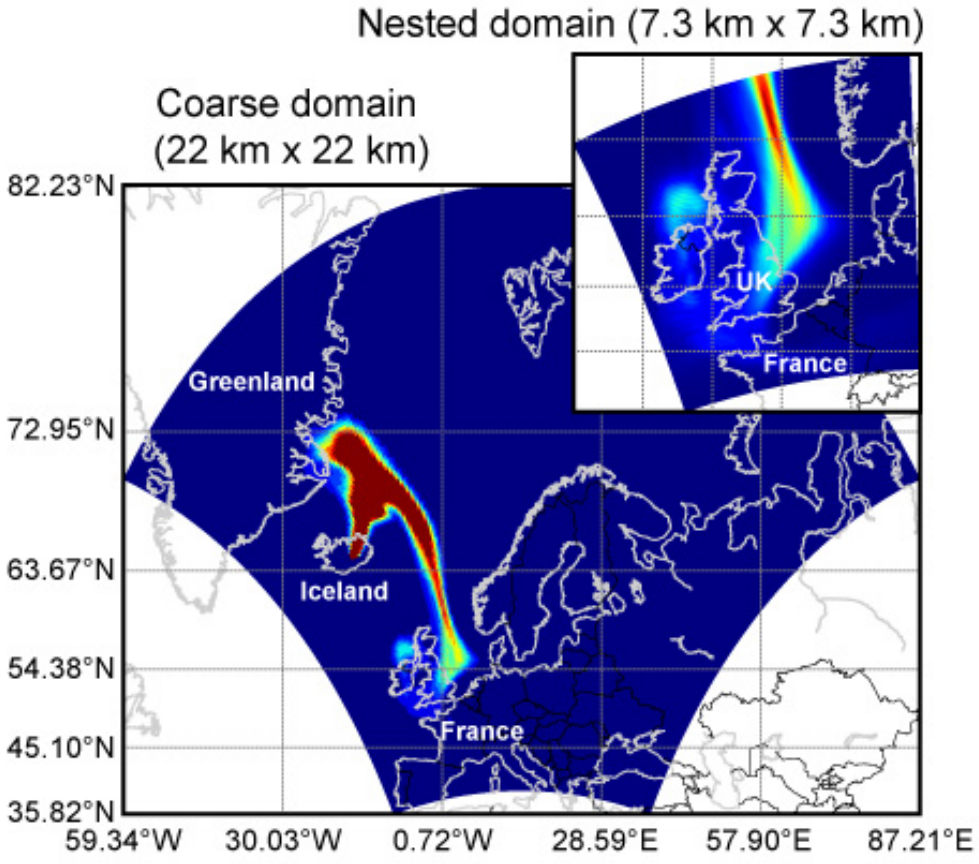

Figure 10. Nested horizontal domains of high spatial resolution simulations: coarse resolution (22 km $\times$ 22 km) and nested high resolution (7.3 km $\times$ 7.3 km) (in inset) domains.

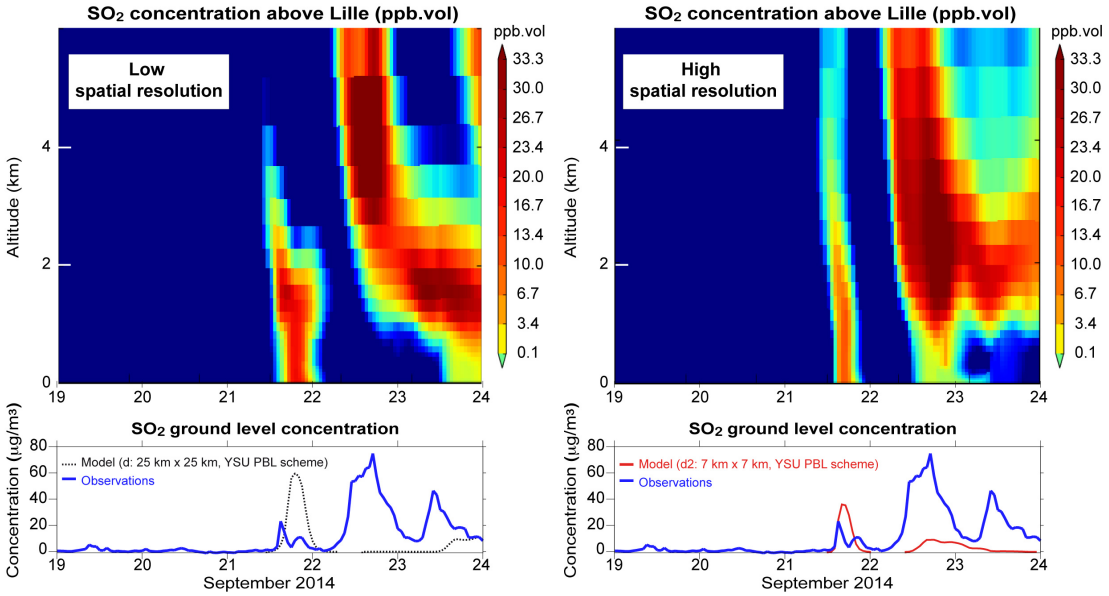

Figure 11. Comparison of far-range vertical distributions of SO_2 over Lille modeled with simulations at (left) low versus (right) high horizontal resolution (both configured with the YSU PBL parameterization scheme). Simulations at low resolution (25 km × 25 km) are performed on a single domain while simulations at higher resolution are performed on two nested domains: the largest with a coarse resolution of 22 km × 22 km and the narrowest with a fine resolution of 7.3 km × 7.3 km (Fig. 10). (Top) Modeled concentration of volcanic SO_2 over Lille as a function of time (X-axis) and altitude (Y-axis). (Bottom) Time evolution of the observed (blue) and modeled (red or black) SO_2 concentrations at ground-level.

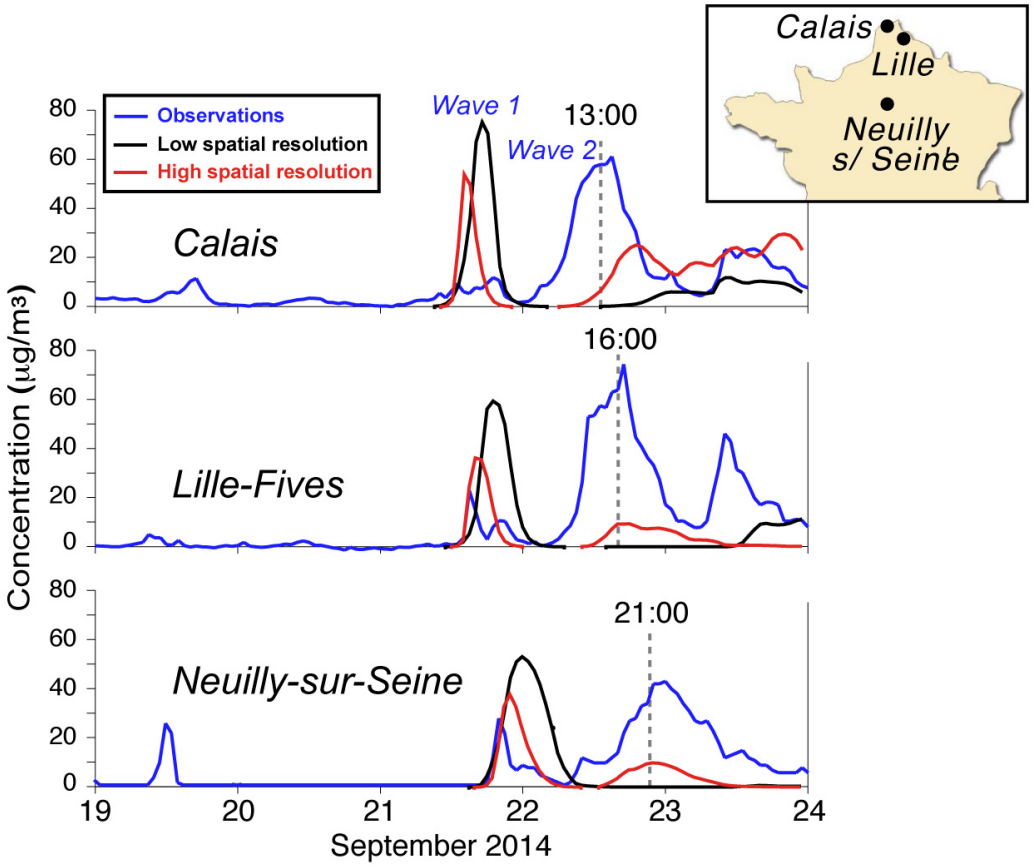

Figure 12. SO_2 ground-level concentration observed by air quality networks in France (blue) and modelled with high (red) or low (black) horizontal resolution simulations, both configured with the YSU PBL parameterization scheme.

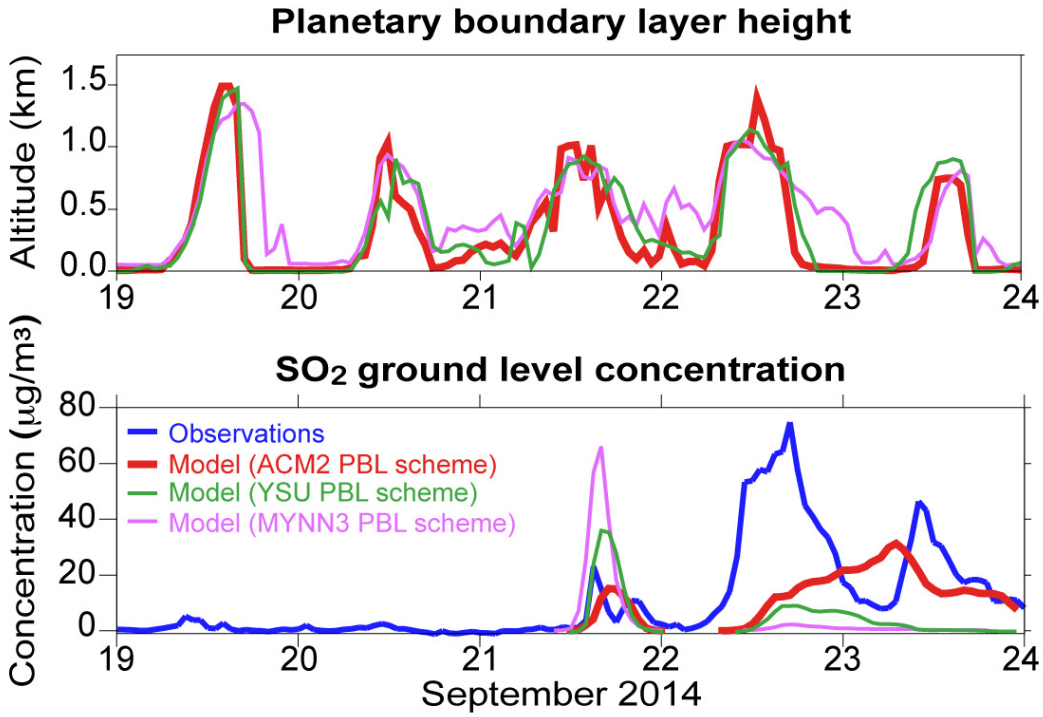

Figure 13. Time variations in Lille of modeled (Top) planetary boundary layer height and (Bottom) SO_2 concentration at ground-level with high spatial resolution simulations configured with YSU (green), ACM2 (red) and MYNN3 (pink) PBL parameterization schemes. The observed ground-level concentration is represented in blue.

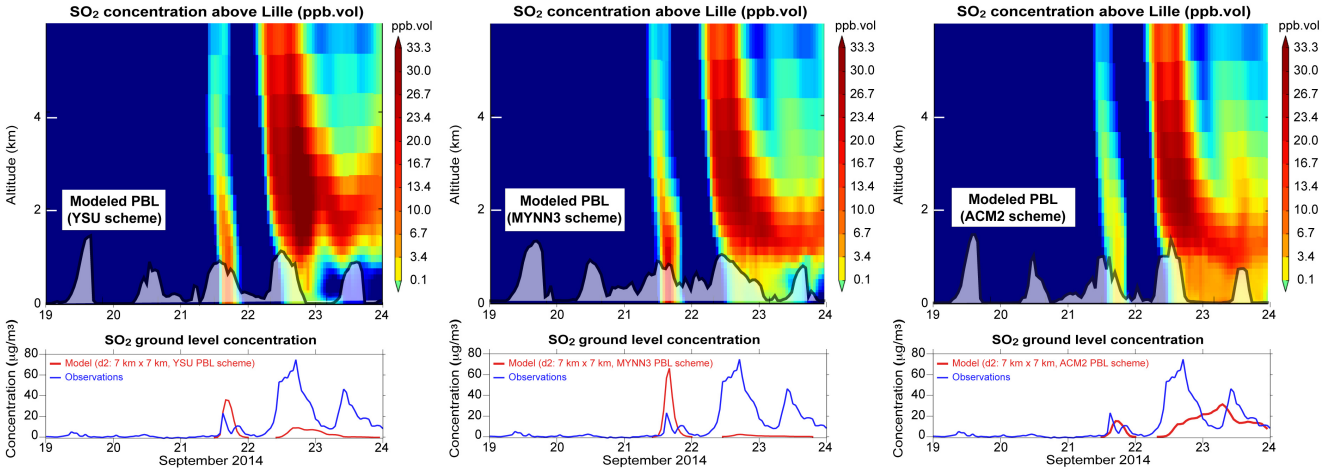

Figure 14. Comparison of far-range vertical distributions of SO_2 over Lille modeled with simulations at high spatial resolution configured with (left) YSU, (middle) MYNN3 and (right) ACM2 parameterization schemes of the planetary boundary layer. (Top) Modeled concentration of volcanic SO_2 as a function of time (X-axis) and altitude (Y-axis). The modeled PBL is overlaid in black. (Bottom) Time evolution of the observed (blue) and modeled (red) SO_2 concentrations at ground-level.

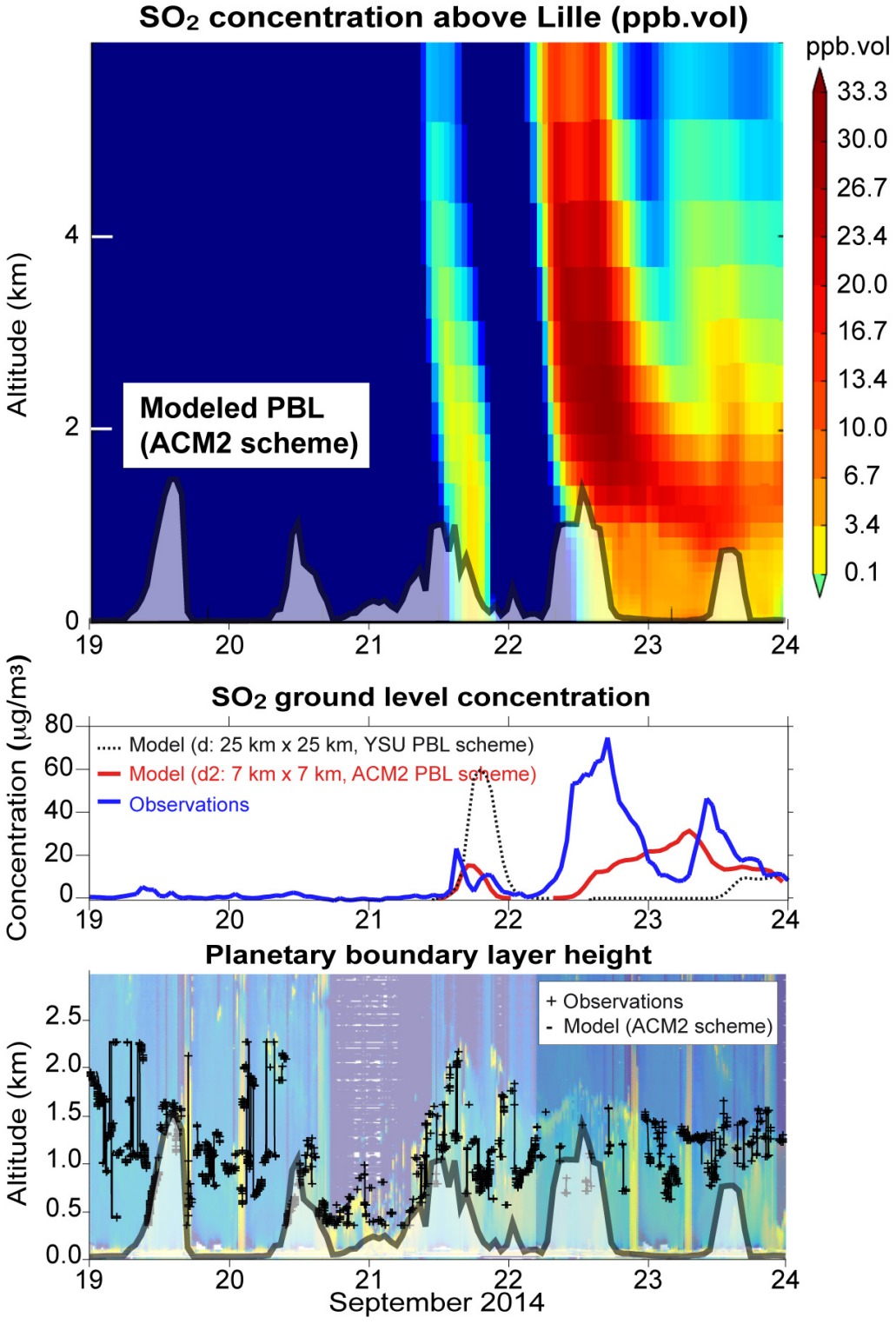

Figure 15. Key role of the PBL dynamics on far-range SO_2 concentrations at ground-level in Lille. (Top) Modeled concentration of volcanic SO_2 as a function of time (X-axis) and altitude (Y-axis) with simulations at high spatial resolution configured with ACM2 PBL parameterization scheme. The modeled PBL is overlaid in black. (Middle) Time evolution of the observed (blue) and modeled (red) SO_2 concentrations at ground-level. (Bottom) Comparison of modeled (black line) and observed (crosses) PBL height with time overlaid on lidar backscatter profile.