

## Response to referee #1

### Specific comments

My major concern is with the relationships drawn between the brightness temperature difference ( $\Delta BT$ ), Absorbing Aerosol Index (AAI) and SO<sub>2</sub> total column amounts (i.e. Fig. 5). By eye, it looks like there is no correlation at all. However, it's difficult to tell as no statistical metrics are given. I suggest adding a statistical metric (perhaps a correlation coefficient if the relationship is expected to be linear) to demonstrate that there's a notable relationship (as the authors claim). Further comment on this is provided in the Technical corrections section.

- *there is indeed no clear relation between  $\Delta BT$ , AAI, and SO<sub>2</sub>, which is an important aspect of the paper. If anything, our results suggest that the relation is complex because the observations were made shortly after the eruption, and the presence of ash and condensed water. We find indications that the presence of ash and condensed water causes a possible shielding effect, i.e. TROPOMI only observes some aspects of the volcanic plume, while the presence of condensed water also hampers the IR  $\Delta BT$  detection of volcanic ash (a known problem, but nevertheless problematic if these type of measurements are to be used in for example aviation applications).*

*with the help of the referee comments and changes we made we think this should be clearer, see further in the responses to the referee comments.*

Another concern is the reliance and interpretation of the 'VADUGS' algorithm. The authors refer to a conference talk, which in general is fine, but as some of the conclusions reached rely on an understanding of this algorithm and its uncertainties a reference to a published article describing it is necessary (in my opinion). If it is not published elsewhere, then a section describing the algorithm should be added if it is to be used in the comparison of the TROPOMI data.

- *This is a sensitive and difficult point. We currently do not have the capacity nor the time to extensively describe VADUGS, which in essence would be a DLR task, as VADUGS is their algorithm.*

*Originally we expected that VADUGS – which has been an operational algorithm at DLR since 2013 – would be described in a separate paper independent of the EUNADICS-AV project by DLR and by the time our paper was submitted. Due to several staffing changes at DLR (including the DLR co-author moving to EUMETSAT) this has not yet happened.*

*However, we would argue that the paper actually does not rely that much on VADUGS but rather mostly on HIMAWARI-8 IR  $\Delta BT$  differences, which is identical for any volcanic ash retrieval algorithm. We do use the retrieved VADUGS heights (but not ash amounts, not effective radius), but only in a qualitative sense.*

*As the reliance on specific and quantitative VADUGS results is limited, the lack of official VADUGS description should not have to be considered as overly important.*

*However, so this is to decide by the editor. And we reiterate that unfortunately there is little we can do to resolve this point by providing a full reference to the VADUGS algorithm.*

Another issue is the interpretation of the CALIOP data. The CALIOP pass clearly showed a feature that reached 18 km (asl). This is not mentioned anywhere. In addition, the feature (on average)

reached cloud-top heights of 16 km (the authors cite a height of 15 km). It is important to get this right as this paper could be a nice reference for the eruption height of the 19 February 2018 Sinabung cloud in the future.

- *we have added the observation that there also appears to be (some) ash up to 18 km, and explained why this layer likely is also of volcanic origin as there are no indications from HIMAWARI-8 that there are other high altitude clouds at those locations at that time. This does not impact the findings of the analysis, as this layer coincides with the thinner part of the volcanic plume for which we argued the TROPOMI height retrievals are less accurate as TROPOMI “sees” partly through the cloud.*

In general, the authors use the terms ash plume, volcanic cloud, volcanic ash, volcanic ash plume, aerosols etc interchangeably to refer to the eruption cloud (and in some cases to refer to components of the volcanic cloud that were ice-rich). I suggest that the authors define these terms early on in the manuscript. This will avoid confusion, especially when discussing the microphysical make-up of the volcanic cloud. One suggestion could be to use the generic term ‘volcanic cloud’ to refer to a cloud of volcanic origin and use the terms ice-rich, ash-rich and SO<sub>2</sub>-rich to refer to regions of the volcanic cloud that exhibit these spectral signatures.

- *we have changed all references to “volcanic ash plume” into “volcanic ash cloud”, all references to “volcanic ash height” into “volcanic ash cloud height”. This leaves essentially two expressions: “volcanic ash cloud (height)”, which refers to the volcanic eruption cloud phenomenon, and “volcanic ash”, which refers to the physical quantity. We checked the document to ensure that “volcanic ash” is only used when we refer to volcanic ash as the physical quantity.*



“ ... we use total attenuated backscatter data from one CALIPSO orbit (data version 3.40) in a qualitative approach, *i.e.* detection of cloud and aerosol layers and their heights.”

P5L158: 'local time of 06:25 UTC' - Is this local time or UTC?

- *'local' here refers to the geographical location, as measurement times change during a TROPOMI orbit. This is indeed confusing, so we changed the sentence to:*

*“with TROPOMI measurements within the figure area made at approximately 06:25 UTC”*

P6L164: 'Cook et al. (2014)'. Could add references to Moxnes et al. (2014) and Prata et al. (2017), which both specifically investigate the separation mechanisms of volcanic ash and SO<sub>2</sub>

- *references added*

P6L166-168: I would consider moving the VIIRS and NOAA/CIMSS volcanic ash retrieval Supplementary Figures into the main manuscript. The true colour VIIRS image is important for context and interpretation of the TROPOMI height retrievals (presented in Fig. 1). Also, use of NOAA/CIMSS retrievals (which are referred to for the cloud height in this sentence) should be stated with the correct references (*i.e.* Pavolonis et al. 2015a, b)

- *this is a point of contention: originally the VIIRS and NOAA/CIMSS retrievals actually were in the main paper. However, as (1) the paper is already rich in figures and (2) the main topic of the paper is evaluating TROPOMI and HIMAWARI-8 volcanic ash heights, we decided at the finalizing stage of writing to move the VIIRS/NOAA/CIMSS results to the SI.*

*Furthermore, in essence the VIIRS/NOAA/CIMSS results are based on the same type of measurements (broad band IR) and the same retrieval approaches (split-channel) as the HIMAWARI- data. However, data files of the VIIRS/NOAA/CIMSS results are not publicly available and accessible, which would allow for direct comparison. Hence, use of results that are only available as imagery, not as data, and we prefer not to rely just on imagery in the main body of the paper.*

*Unless it is really crucial for the paper – which believe it is not - we would prefer to keep the VIIRS and NOAA/CIMSS figures in the SI.*

- *references have been adjusted as suggested*

P6L171: 'Systematically higher' - This implies FRESCO cloud heights are always higher than the O22CLD heights. Based on Fig. 3, this looks to be the case from 3-5 degrees latitude. However, from 2-3 degrees latitude it looks like O22CLD is higher than FRESCO. So, I wouldn't call this systematic. Perhaps it would be simpler to state 'In general, FRESCO cloud heights are higher than the O22CLD heights'. Or something similar. A correlation plot could also be added to show the bias of FRESCO/ROCINN vs. O22CLD cloud heights.

- *changed sentence as suggested*
- *For general interest: we did a check: for the region of the plot, and for 88% of clouds with CTH > 5 km, FRESCO CTH are higher than O2O2 CTH. For a wider geographical area (entire orbit), this drops to 60%. However, when instead considering clouds with CTH > 10 km for the*

*wider area, this fraction increases again, even further for larger cloud fractions (0-25-50-75% cloud fraction 67-71-76-87%). For ROCINN, these numbers are even better (89-91-93-97%).*

P6L176: 'up to 15 km altitude' - Please provide a reference for this. Also, how strict is this limit? In Fig. 3, I see cloud heights higher than 15 km. Also, is this above sea level? Please make this clear in the text.

- *changed to:*

*"up to approximately 17 km altitude (~100 hPa) [Wang et al., 2012]"*

- *This is a typo, it should read "25 km", which is a hard-coded limit for FRESCO CTH. However, a more realistic limit – and hence why we overlooked it, is that in reality FRESCO has – by virtue of its oxygen absorption – an acceptable sensitivity up to approximately 100 hPa or approximately 17 km altitude. Research papers on both FRESCO validation as well as FRESCO methodological uncertainties indicate suggest an uncertainty range of 25-50 hPa. Furthermore, there is limited information about the quality of FRESCO CTH above 15 km there are few validation opportunities. Clouds with clouds top heights at 15 km or above are rare, even more so optically thick clouds for which the FRESCO "centroid altitude" lies at 15 km or above. However, with increased possibilities for measuring clouds above 15 km altitude (more & better satellites), as well as interest in high altitude clouds above 15 km altitude, this will be a valuable future research topic.*

P6L183: Please provide a colour scale/legend with Fig. 2 to show which AAI values correspond to which colour.

- *a color scale was added*

P6L183: Interpretation of the CALIOP data. Based on Fig. 2, it looks like the main feature has cloud-top heights of around 16 km (15 km is stated in the manuscript). There is also a clear feature at 18 km (detected by the AAI). This is not mentioned at all and should be addressed in the manuscript.

- *changed to "approximately 16 km"*
- *added a short discussion about this 18 km plume later in section 3.4 in relation to Figure 4.*

*"There is also a layer detected in CALIPSO at 18 km around 3°N, which likely is also volcanic as the HIMAWARI-8 BT does not provide any indication of other high clouds while there are negative ΔBTs near the CALIPSO track at 3°N, indicative of the presence of volcanic ash."*

P6L188-189: There is poor agreement between FRESCO and CALIOP from 3-4 degrees latitude, which should be stated here.

- *changed to:*

*"Between 3° and 4° latitude, the agreement is poor as the FRESCO"*

P7L193: CALIOP's feature mask - Please state which version of the feature mask is being interpreted. There were changes made from V3 to V4. I looked at the VFM V4 for this pass and I can see some small parts of the feature classified as dust aerosol but the majority is cloud.

- *added, it is v3.4*

P7L194: 'clearly the attenuation is not complete.' - I'm not sure it is that clear. This interpretation would be more justified if the VFM was plotted on the same scale as Fig. 2 and inserted as a second panel.

- *correct, although even for CALIPSO v4.10 the nearly all cloud pixels for this scene are flagged as "cloud". We changed the sentence to:*

*"does not identify hardly any of these backscatter signals as aerosol (for CALIOP v4.10 an occasional cloud pixel is flagged as aerosol):"*

P7L196: Comparison of FRESCO and ROCINN – is this only for AAI > 0? Please clarify.

- *The R2 = 0.98 is based on all clouds regardless of AAI value. Sentence changed to:*

*"FRESCO cloud heights between 0.5 and 14 km regardless of corresponding AAI value"*

P7L200-201: 'and all data products increasing heights in the volcanic cloud going from south to north.' - This is simply not true. The heights increase from 2-4 degrees latitude and then decrease from 4-6 degrees latitude. Please clarify in the text.

- *This is sloppy formulation from our side, the point we wanted to make – and the change we made - is:*

*"... with the largest heights between 4° and 5° latitude, consistent with the CALIOP observation that backscatter signals between 3° and 4° latitude are weaker than between 4° and 5° latitude"*

P7L209-210: 'The eruption dynamics may thus have additional effects on the ash plume displacement, but this cannot be investigated based on the available satellite data.' - This statement requires further justification and clarification about why the available satellite data cannot be used to study the eruption dynamics. For example, Himawari-8 provides excellent observations (every 10 minutes) of the volcanic cloud's evolution and dynamics (as the authors discuss in Section 3.4).

- *What would be needed for properly understanding the eruption dynamics are time series of its 3D structure (time-lon-lat-height), like in a model, but which clearly cannot be observed or reconstructed from satellite observations. Satellites thus only provide a partial view of the entire eruption plume.*

*Changed the description to:*

*"The eruption dynamics may thus have additional effects on the ash plume displacement, for which time series of the complete 3-dimensional view of the eruption plume would be preferred. The current available satellite data only provide a 2-dimensional view of the eruption plume from above (geostationary, Polar orbiting), with information about changes over time in case of the geostationary satellites and with some but limited information about cloud and aerosol height. CALIOP measurements only provide one 2-dimensional cross-section through the eruption plume, without any information about changes over time."*

P7L212: Change 'is' to 'was'.

- *changed*

P7L216: Positive  $\Delta BT$ s are also indicative of clouds composed of water droplets (not just ice). Please clarify in the text.

- *changed to:*

*“with negative  $\Delta BT$  potentially indicating volcanic ash, and positive  $\Delta BT$ s indicative of the presence of nontrivial liquid water or ice content [Pavolonis et al., 2006].”*

P7L218-219: ‘one associated also with a high cloud height, and another one further south with much lower cloud heights’ - what cloud heights are being referred to here? The VADUGS algorithm in Fig. 4 only appears to show a high altitude cloud.

- *some reddish colors within the contours, potentially indicating volcanic ash, collocated with high clouds (whites), while other reddish colors collocated with low clouds (blues). To describe exactly what we note, we changes the sentence to:*

*“one associated also with a high cloud height (white cloud colors), and another one further south with much lower cloud heights, likely low-altitude outflow or pyroclastic flows (blue cloud colors).”*

P7L221: ‘dense high ice clouds’ - What do the authors mean by ‘dense’ here? Optically thick ice clouds would show a near zero  $\Delta BT$ , not a strongly positive  $\Delta BT$ .

- *dense refers to the observations that initially the high cirrus is not transparent (see also SI figures S 2A/2B). It is not critical, so we modified the text in combination with the next comment.*

P7L221: ‘purple region’ - I actually see this as blue. Maybe call it an ‘ice-rich cloud’?

- *see also previous comment, sentences were changed, the color purple now only refers to the  $\Delta BT$ s:*

*“large positive  $\Delta BT$ s (purple), indicative of high ice clouds, which continues to grow and expand northward.”*

P7L222-224: Figure 5 - I found this figure difficult to interpret. At this line in the manuscript the authors refer to the ‘HIMAWARI VADUGS  $\Delta BT$ s’. How are VADUGS  $\Delta BT$ s different to a simple 11-12 micron  $\Delta BT$ ? In Fig. 5 they just look like  $\Delta BT$ s. The authors also state that ‘When focusing on AAI and SO<sub>2</sub> values, it appears that larger  $\Delta BT$  values occur for smaller AAI values (< 2) and SO<sub>2</sub> (< 20 DU)’ - For the lower left plot in Fig. 5, I can see numerous data points that have positive  $\Delta BT$  (0-10 K) for large (2-6) positive AAI values (contradictory to what the authors claim) and I find it very hard to interpret any relationship whatsoever in this panel. In Fig. 5 lower right panel, again, it’s hard to see any relationship because there are positive and negative  $\Delta BT$  values that correspond to a whole range of SO<sub>2</sub> values (5-100 DU).

There are several ways Fig. 5 could be improved: First, I would only plot the data that falls within the contours plotted in the upper left panel of Fig. 5 as this clearly contains the volcanic cloud (what are these contours by the way? They are not mentioned in the Fig. 5 caption). This would remove the black dots (I assume?), which at the moment are distracting. Second, some kind of statistical metric

could be used to indicate that there is indeed a relationship between AAI,  $\Delta BT$  and  $SO_2$ . If the relationship is not linear then maybe some kind of curve fit (exponential for lower right panel?) will help the reader interpret the relationships.

- *'HIMAWARI VADUGS  $\Delta BT$ s' should read 'HIMAWARI  $\Delta BT$ s'.*
- *The point of the comparison between  $\Delta BT$  on the one hand and the AAI and  $SO_2$  on the other hand is to check if there was any relation between both. Based on passed literature showing that the magnitude of  $\Delta BT$  is not related to ash optical depth (Figure 2 of [Prata & Prata, 2012] provides a nice illustration of this point), the "naive" assumption would be that  $\Delta BT$  then should also not show a clear relation with the AAI and  $SO_2$ . Which is doesn't, as the largest AAI and  $SO_2$  values occur for smaller  $\Delta BT$ s. This is in a way a "negative" result, lack of a relationship between two parameters, but we felt that it was important to show this, as it highlights our point that there is added value in combining IR  $\Delta BT$  with UV/VIS AAI and  $SO_2$ .*

*We added the following sentence to this section:*

*"Overall, we find little evidence of large AAI values and large  $SO_2$  values associated with large  $\Delta BT$ s. Rather, their relation is complex."*

- *In addition, we also added the following to the discussion & conclusion section (added section underlined here):*

*"... synergistically combining different satellite data products like the AAI and  $SO_2$ . Furthermore,  $\Delta BT$  appears not to be a good indicator of either large AAI values or large  $SO_2$  columns. This is not surprising as  $\Delta BT$  is not a good indicator for ash optical depth [e.g. Prata and Prata, 2012; Pavolonis et al., 2016]. Our results therefore highlight that there is added value in combining IR  $\Delta BT$  with UV/VIS AAI and  $SO_2$ . Satellite measurements ... "*

- *The solid and dotted contours denote outline of TROPOMI > 10 DU  $SO_2$  columns and TROPOMI AAI > 0 value. This information is provided in the figure caption of figure 4, which the figure caption of figure 5 refers to. We added the same explanation to the caption of figure 5.*
- *Figure 5 is modified with now only showing three panels (3x1 rather than 2x2), with in the spatial regridded  $\Delta BT$ s only the  $\Delta BT$ s within the  $SO_2$  /AAI contours (see here below for explanation of the contours). The lower two plots have remained. Color bars have been added. The figure caption now reads:*

**"Figure 5. (A)** HIMAWARI-8  $\Delta BT$ s for 19 February 2018 06:30 UTC (see also Figure 4) regridded to the TROPOMI measurement grid of that day, and correlations between the HIMAWARI-8  $\Delta BT$ s and TROPOMI **(B)** AAI and **(C)**  $SO_2$ . The solid and dotted contours denote outline of TROPOMI > 10 DU  $SO_2$  columns and TROPOMI AAI > 0 value, as also shown in figure 4 and derived from Figure 1. The color coding of the dots in the AAI scatterplots is indicative of the corresponding  $SO_2$  value (> 10 DU) , and the color coding in the  $SO_2$  scatterplot is indicative of the AAI value (AAI > 2), see also the lower color bar. These color codings were added for qualitatively identifying possible relationships between  $\Delta BT$  and AAI or  $SO_2$  within the volcanic ash plume."

P7L224: 'The larger  $\Delta BT$  are also associated with optically more dense clouds (see VIIRS imagery in the SI and comparison of TROPOMI with CALIPSO).' - This statement needs to be further clarified. It's

not physically possible for an optically thick cloud to have a large  $\Delta BT$  in the infrared. When clouds become optically thick they behave as grey bodies (little spectral variation across thermal infrared wavelengths) and so a difference in brightness temperature between 11 and 12 micron should be close to zero. However, I think what the authors are observing is a relationship between high reflectance at visible wavelengths (white clouds in VIIRS imagery) and large  $\Delta BT$ s, but it's not clear in the way that it's stated.

- *We agree, the comparison with  $\Delta BT$  and  $SO_2$  /AAI requires careful wording. The main points we want to make is that the relationship between all three parameters is complex, and that we find indications of possible shielding effects (thick ash/ice shielding part of the underlying ash/ $SO_2$  column). We modified this section as follows:*

“Figure 5 shows a comparison of TROPOMI AAI and  $SO_2$  data with regrided HIMAWARI-8  $\Delta BT$ s (upper left plot). When focusing on AAI and  $SO_2$  values, it appears that larger  $\Delta BT$  values occur for smaller AAI values ( $< 2$ ) and  $SO_2$  columns ( $< 20$  DU). The largest positive  $\Delta BT$  are associated with optically thicker/less transparent water and ice clouds (see also VIIRS imagery in the SI and comparison of TROPOMI with CALIPSO). The lack of larger AAI and  $SO_2$  values for larger positive  $\Delta BT$  values therefore may reflect some kind of shielding of the volcanic ash and  $SO_2$  by the iced upper levels of the volcanic ash cloud.  $SO_2$  may have been converted into sulphate as the SO depletion rate (e-folding time), which, although uncertain, has been estimated to be as small as 5-30 minutes [Oppenheimer et al., 1998; McGonigle et al., 2004], scavenged by ice [Rose et al., 2000], or via ice nucleation of volcanic ash particles [Durant et al., 2008]. For negative  $\Delta BT$ s – indicative of volcanic ash – we also find little evidence of a distinctive relation between either the AAI and  $SO_2$  with  $\Delta BT$ s. This may similarly reflect a shielding effect, as the largest  $\Delta BT$ s do not occur for the largest aerosol concentrations [e.g. Prata and Prata, 2012; Pavolonis et al., 2016].”

P8L225-227: This could be due to scavenging of  $SO_2$  by ice (Rose et al., 2000). It could also be due to ice nucleation of volcanic ash particles (Durant et al., 2008). In terms of the conversion of  $SO_2$  to sulphate, is there a reference that could be added here? i.e. how long does it typically take for  $SO_2$  to convert to sulphate in the upper troposphere? And does this conversion rate make sense given the time of observation and time since eruption?

- *Mechanisms and references included in the text (see previous question for text modifications). The  $SO_2$ -to-sulphate conversion time scale (e-folding time) is highly uncertain but can has been estimated as small as 5-30 minutes. Both remarks – including references – have also been added (again, see previous question for the new text).*

P8L245-248: ‘Comparison with geostationary IR volcanic ash height’ - Which retrieval is this statement referring to? Is this the VADUGS volcanic ash cloud height retrieval? It's the comparison with CALIOP that demonstrates TROPOMI height algorithms may underestimate heights for semi-transparent ash clouds. Please clarify this.

- *Should be CALIOP, not geostationary IR. This has been changed.*

P8L251-252: The ‘shielding’ effect - This is rather speculative and could be due to a number of different reasons (see previous comments on P8L225-227). Also, is this shielding of  $SO_2$  or ash or both? I think to substantiate this claim, evidence of  $SO_2$ /ash existing underneath the cloud-top should be provided.

- *As explained, the lack of a distinctive relation between  $\Delta BTs$  (both positive and negative) on the one hand and AAI and  $SO_2$  on the other hand we interpret as indications for shielding effects, see further the answer to P7L224. There does not exist other data to substantiate this claim. However, with the mentioning of other processes like  $SO_2$  conversion, depletion, scavenging, and nucleation, it should be clear to the reader that the analysis is by far definitive with regarding to the presence of a shielding effect.*

P9L257-258: 'the retrieval algorithm' - which retrieval algorithm is being referred to here? Please clarify.

- *Should read "IR volcanic ash retrieval algorithms", changed accordingly.*

P9L266-268: 'TROPOMI cloud heights can be used for determining aerosol heights for AAI values greater than 4' - How was this conclusion reached? What is the significance of  $AAI > 4$ . As stated in the previous sentence, the TROPOMI cloud heights do not perform well for semi-transparent clouds regardless of their AAI value. This statement requires further clarification. Also 'column values  $> 1$  DU' is TROPOMI's signal-to-noise really this good? Please provide a reference.

- *preamble: although the AAI is not a quantitative parameter, in general there is – all else being equal - a clear correlation between the AAI and the AOD. The problem here is the "all else being equal": the AAI value depends on many parameters, like aerosol type, aerosol height, solar zenith angle, viewing angle, albedo below the aerosol layer (which includes clouds), and some more [de Graaf et al., 2005].*

*A large AAI value will – generally speaking – indicate thick aerosol layers. Both user experience and theoretical calculations suggest – again, generally speaking – that AAI values larger than 2 (two) indicate significant amounts of aerosols. To be on the safe side and because the main interest is also to have non-transparent aerosol layers, we increased the AAI threshold to 4 (four). According to de Graaf et al. [2005], for aerosol layers with AAI values  $> 4$ , the AOD generally will be (much) larger than one (AOD of  $1 = 1/e$  or 36.7% of the light is not scattered by the aerosol layer). At such scattering levels, aerosol layers become opaque.*

*We added the following sentence:*

*"This AAI threshold value of 4 may be conservative but ensures that the aerosol layer very likely is opaque, as generally the associated aerosol optical depth will be (much) larger than on [de Graaf et al., 2005]."*

- *TROPOMI  $SO_2$  accuracy is estimated to be at least 1 DU, but likely even better, see Theys et al. [2017]. Reference added.*

P11L323-325: Please fix reference formatting here. Also link provided to Stein Zweers (2016) results in a 'Page not found' error.

- *changed (document recently officially accepted by ESA, so the filename changed)*

P16L411: Check style for figure labels e.g. 'A+E' should be '(a) and (e)'.

- *checked and updated*

P19L415: VADUGS cloud heights are on the right column of Fig. 4 not left and the  $\Delta$ BTs are on the left.

- *changed*

P19L427: Change 'derived' to 'shown'.

- *changed*

P19L427: What is the  $\Delta$ BT bias correction? This needs to be explained and defined in the manuscript.

- *This refers to a typical value of the atmospheric correction of geostationary infrared brightness temperatures for this part of the world, but I do not know why this remark – stemming from an email exchange - has managed to seep into the paper. This can be removed.*