

Response to reviewer comments: Effect of volcanic emissions on clouds during the 2008 and 2018 Kilauea degassing events

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Review Reports

The authors thank the reviewers for the thorough comments. Responses are given below.

Reviewer 2 comments and authors' responses

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General comments:

1. **Comment:** *What is the actual contribution of this study to previous works exploiting volcanic emission to characterize the impact of aerosols on ice and liquid cloud processes? Some indications are given in text, but I suggest to clearly justify the rationales for this study in Introduction and to underline the contributions to previous works in the Abstract and the Conclusion.*

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Response: The following text has been added to clarify our contribution in the context of previous work:

– Abstract:

Although previous studies have assessed the modulation of cloud properties during the 2008 event, this is the first time such an analysis has been reported for the 2018 event and that multiple degassing events have been analyzed and compared at this location.

15

– Introduction:

The 2008 and 2018 degassing events of Kilauea Volcano have been previously studied with respect to characterization of volcanic and seismic activity during peak emissions (Nadeau et al., 2015; Neal et al., 2019; Elias and Sutton, 2012; Wilson et al., 2008; Orr and Patrick, 2009; Orr et al., 2013; Patrick et al., 2019), analysis of 2008 satellite observational data (Beirle et al., 2014; Mace and Abernathy, 2016; Eguchi et al., 2011; Yuan et al., 2011), comparison of 2008 observed and simulated anomalies (Malavelle et al., 2017), and analysis of the effects of 2018 Kilauea emissions on air quality (Tang et al., 2020).

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– Conclusion:

Previous analyses have characterized the geologic and atmospheric effects of the 2008 event, but this is
the first time that such an analysis has been performed for the 2018 event.

- 25
2. **Comment:** *What are the scientific motivations for selecting the Kilauea volcano? I understand that the clean environment allows to separate the impacts of sulfate and ash from other confounding emission sources. Is it the only site offering such “clean” conditions ?*

Response: Not only are local anthropogenic emissions low in the local environment surrounding Kilauea, but because
the island is situated on an island approximately 1000 miles from the nearest major anthropogenic emissions source
(U.S. West coast), there is a low likelihood of multiple emission sources of similar magnitude. For example, Malavelle
et al. (2017) performed a similar analysis of volcanic emissions from a volcano in Holuhraun, Iceland, but could not
completely partition volcanic effects from anthropogenic emissions in Western Europe carried by seasonal wind currents.
Additionally, Kilauea is well-studied and has had long-term monitoring of emissions, seismic activity, and eruptive
behavior over the historical record. Kilauea is a low-altitude volcano, so aerosol injection height is controlled by the
strength of the eruption, which provides us with an opportunity to study aerosol-cloud interactions for liquid and ice
cloud phases separately. Finally, we clearly take advantage of the unique opportunity of analyzing two events around
the same location but with different magnitude in emissions and plume heights. We have clarified this information in the
introduction.

- 30
3. **Comment:** *The authors have chosen to have the results and the discussions in the same section. This is a possible structure for the paper. However in this current form, the result interpretation and discussion are sometimes not enough developed. The back and forth between result description and their interpretation makes the reading difficult (e.g. sub-sections 5.0.1 and 5.02).*

Response: Thank you. We have made an effort to reorganize these sections to increase clarity. Please see detailed
responses to comments below.

- 35
4. **Comment:** *Finally, the limitations of this work need to be acknowledged. Particularly, the uncertainties in the observations used for the analysis have not been discussed or acknowledged properly when comparing satellite retrievals with simulated cloud properties. The discussion does not provide enough insights on the possible shortcomings in the modelling of aerosol-cloud interaction. Recommendations for model improvement could be further developed in the Conclusion.*

Response: We have updated the text to highlight limitations of data products and explicitly cited references. In Section 3 (Data), we point the reader to cited references for uncertainty quantification of satellite retrievals for MODIS variables
as

55 Uncertainty of MODIS retrievals is discussed in Hubanks et al. (2015) and Bennartz (2007); Bennartz and Rausch (2017) (latter is for CDNC only) . . .

, for CALIPSO as

For each level, the gridbox fraction of cloudy, clear, and uncertain areas sum to 1, with the JJA seasonal mean uncertainty (2006–2008) ≤ 0.05 above the boundary layer (Chepfer et al., 2010) . . .

, and OMI

60 For uncertainty quantification of OMI retrievals, see Carn et al. (2016, 2017); Yang (2017).

Furthermore, partitioning of MODIS cloud phase is discussed in Section 7.1:

Another reason behind the larger aerosol effects on COD and LWP simulated by GEOS than observed by MODIS may lie in differences in phase partitioning (i.e., liquid vs. ice) (Marchant et al., 2016). To help identify thin cirrus clouds, measured top-of-the-atmosphere (TOA) reflectance at 1.38 μm is used to partition
65 high-altitude cirrus clouds from low-altitude liquid clouds. This method is strongly influenced by the relative humidity of the atmospheric column. Therefore, areas with low column water vapor amount may have more clouds partitioned to ice phase than is realistic. This is important because 2008 was a La Niña year and the relative humidity in the atmospheric column was lower than normal, so LWP anomalies may appear low because more clouds were partitioned to the ice phase in the MODIS cloud phase classification algorithm.
70 Even in 2018, most of the total water path (TWP) . . .

5. Introduction

– **Comment:** *Introduction, line 20 page 1 : The authors could give some figures illustrating the uncertainties in the radiative impact of ACI (IPCC report). It should be clearly stated in this first paragraph that the understanding of and the representation of ACI in current models represent a major source of uncertainties for NWP and climate studies (I can see that this is developed in the subsequent paragraph)*
75

Response: While we agree that it is important to acknowledge GCM uncertainty, we do not feel that another figure would add significant value to this work, given that the suggested figure applies only to a small portion of the introduction and the IPCC report is cited several times within the introduction in reference to GCM uncertainty. However, we have added the following text to the first paragraph of the introduction to summarize topics that are
80 introduced more fully throughout the text:

Additionally, cloud formation is a complex and nuanced physical process occurring on scales far smaller than those resolved by climate models, and the precise feedback mechanisms influencing AIEs on various timescales are not fully understood (Boucher et al., 2013; Klein et al., 2013; Malavelle et al., 2017; Yuan et al., 2011).

85 – **Comment:** *Introduction, page 3: I suggest that the authors provide more rationale on using the Kilauea degassing event compared to other volcanic events. What is the rationale behind?*

Response: Please see the response to the general comment on site selection.

– **Comment:** *The outline of the paper should be given at the end of Introduction on page 3 this will give a transition with Section 1.*

90 **Response:** We have added a brief outline at the end of the introduction as requested.

6. Section 1

– **Comment:** *The Section 1 which is dedicated to the volcano description is interesting but may be too long. The authors should better emphasize the differences between the 2008 and 2018 events in terms of injection height, degassing composition and amount, type and duration of eruption. . . A table could help.*

95 **Response:** Thank you for pointing this out. We have condensed the description of events at Kilauea into a single subsection that focuses on the amount of SO₂ emissions, plume height and plume composition for each event, as well as any distinguishing characteristics.

7. Section 2

– **Comment:** *Page 5, line 20: It is not clear what processing is applied to missing values, is it gap-filling?*

100 **Response:** Corrected to say: “Missing values in MODIS data, primarily found in CDNC and ice products, were smoothed using cubic spline nearest-neighbor interpolation and a gaussian filter.”

– *the description of the satellite products given in page 5 is not accurate enough.*

– **Comment:** *What are the variables used in the MODIS and CALIPSO cloud products, is it cloud fraction ? optical depth ? Please list here all the retrieved variables used in your results (a table including all the symbols and acronyms could help).*

105 **Response:** Thank you. We defined these parameters in the Results and Discussion. We have defined the acronyms used in Section 2 (data) and specified which datasets were used from each satellite source. We have also added a lookup table describing each variable.

– **Comment:** *What is the vertical resolution of CALIPSO data ?*

110 **Response:** Corrected to say: “. . . and vertical resolution typical for most GCMs (40 levels; $\Delta z = 480$ m)”

– **Comment:** *What is the temporal frequency of the CALIPSO product?*

Response: We used the monthly product. This has been added to the text

– **Comment:** *How the anomalies (shown in Fig 5) have been computed for CALIPSO?*

115 **Response:** They represent the difference between the long term mean (2006-2017, 2008 excluded) and the period of each of the events. This explanation has been added to the text.

- **Comment:** *What are the rationales for using the MODIS and CALIPSO products ? What are the value-added of each product in term of information content for this work ?*

Response: MODIS and CALIPSO are the only instruments that provide retrieved level 3, gridded cloud microphysical properties during the two events, and for which satellite simulators have been developed and implemented. Hence they are essential to validate the modelled cloud properties and the accuracy of the GCM. We have added a paragraph at the beginning of the Section 2 (Data) that introduces the satellite products, which datasets are used, and cites previous works using these products and for which purposes.

- **Comment:** *I would suggest to give some insights on the retrieval algorithm used for each product along with the associated key references (this is partly given for MODIS but missing for CALIPSO)*

Response: We have added the following text to the description of GOCCP data:

Instantaneous profiles of the lidar scattering ratio are computed and used to infer the vertical and horizontal distributions of cloud fraction. For each level, the gridbox fraction of cloudy, clear, and uncertain area sum to 1, with the JJA seasonal mean uncertainty (2006-2008) ≤ 0.05 above the boundary layer (Chepfer et al., 2010).

- **Comment:** *what are the uncertainties associated with the MODIS and CALIPSO products ? Could you provide product evaluation references ?*

Response: The references we have already cited provide this information, we have updated the text to clarify.

8. Section 3

- **Comment:** *page 6 line 5 : Which model is it simulating the advection of the aerosol and trace gases: GOCART of GEOS ? What is the type of transport scheme (e.g. semi-lagrangian ?) Is GOCART a model embedded in the GEOS model ? How are coupled both models ? page 6 line 5-10: A separate paragraph should be dedicated to the aerosol model: type of aerosols, bin size, main simulated processes, key references...This needs to be given before the statements on emission sources.*

Response: The following paragraph was added to Section 3:

Transport of aerosols and gaseous tracers such as CO were simulated using the Goddard Chemistry Aerosol and Radiation model (GOCART) (Colarco et al., 2010), which interactively calculates the transport and evolution of dust, black carbon, organic material, sea salt, and SO₂. Dust and sea salt emissions are prognostic whereas SO₂ and biomass burning and antropogenic emissions of SO₂, BC, and OC are obtained from the Modern Era Retrospective Reanalysis for Research and Applications-Version 2 (MERRA-2) dataset (Randles et al., 2017). GOCART explicitly calculates the chemical conversion of sulfate precursors (dimethylsulfide or DMS, and SO₂) to sulfate. The aging of carbonaceous aerosol is represented by the conversion of hydrophobic to hydrophilic aerosols using a e-folding time of 2 days (Chin et al., 2009). Using the evolving meteorological fields from GEOS, each time step GOCART simulates the advection (using a flux-form semi-Lagrangian method, Lin and Rood (1996)), convection, as

150 well as wet and dry deposition, of aerosol. Aerosol optical depth (AOD) is calculated as a function of
aerosol size distribution, refractive indices, and hygroscopic growth. Each aerosol type is assumed to be
externally mixed. Size distributions are prescribed for different types, using 5 bins for dust and sea salt,
respectively, and single lognormal modes for other aerosol components (Colarco et al., 2010; Chin et al.,
2009). This approach was also used to estimate the aerosol number concentration used in the calculation
155 of aerosol-cloud interactions (Barahona et al., 2014).

– **Comment:** *page 6, line 9-10: How is volcanic SO₂ constrained by OMI data ? (data assimilation ?)*

Response: We have clarified how OMI emissions were used with the following text in Section 3:

Volcanic SO₂ emissions are constrained by observations from the Ozone Monitoring Instrument (OMI)
on-board NASA's EOS/Aura spacecraft (Carn et al., 2015). For Kilauea, this dataset only provides “con-
160 stant” annual SO₂ emission rates. For the 2008 event, we replaced this data set with daily varying emis-
sions (Carn et al., 2017; Yang, 2017). Daily emissions for 2018 were obtained from Li et al. (2020).
Missing values were replaced with Ozone Mapping and Profiling Suite data (<https://so2.gsfc.nasa.gov/>)
whenever possible, otherwise the nearest real data point was used. Vertical column density data were
converted from molecules of SO₂ cm⁻² to kg sulphur per second (kg S s⁻¹) using the linear relationship
165 shown in (Beirle et al., 2014, Fig. 6).

– **Comment:** *page 6, line 11-12: could you be more precise on the daily varying emission data set used in this work
? Is it from OMI data as well ? This is not clear*

Response: Done.

– **Comment:** *Is the cloud microphysics scheme a GEOS component ? What is the name of the scheme ?*

170 **Response:** Cloud microphysics is part of the “moist” component of GEOS, which includes the evolution of strati-
form and convective clouds. The former is described using the Morrison and Gettelman (2008) scheme, sometimes
called “MG1”. The microphysics of convective clouds are described using the scheme developed in Barahona et al.
(2014).

– **Comment:** *Is the GEOS model constrained by data assimilation, particularly for aerosol (MODIS AOD ? ...)*

175 **Response:** Winds and temperature are constrained to the MERRA-2 reanalysis. We run the model in “replay”
mode. This technique can be loosely described as a nudging scheme that minimizes the instability and the numerical
drifting associated with regular nudging techniques (Takacs et al., 2018). However we didn't constraint water vapor
nor aerosol concentrations, even though they are available in MERRA-2. Doing so would have limited the response
of clouds to aerosol emissions (via aerosol activation) and vice-versa the response of aerosols to cloud formation
180 and precipitation (via scavenging), adding great difficulty to the analysis. This explanation has been added to the
text.

– **Comment:** *the last paragraph (page 6 line 25-35) concerns the model implementation. I suggest having a dedicated
subsection 3.2 to model configuration and a subsection 3.1 on general description of the model*

185 **Response:** We reorganized Section 3 (GEOS model description) as follows. The general model description constitutes the main section content (AGCM description, general description of aerosol transport, microphysical scheme), while a subsection (3.1: Experimental configuration) outlines implementation-specific details (emissions sources/modifications, and constraints).

Section 4

– **Comment:** *page 7 line 22 : which retrieval is used here: cloud fraction, AOD ?*

190 **Response:** We obtained level 3 MODIS retrievals of AOD, cloud fraction, effective radius, optical depth, and liquid and ice water path. The Data section has been updated to specify the retrievals used.

9. Section 5

– **Comment:** *page 9, line 8-12: The following findings are missing from the analysis of Figure 2*

195 – **Comment:** *Figure 2 shows a better agreement between the MODIS anomalies and the GEOS anomalies in 2018 compared to 2008. Particularly, the spatial extension of the plume in 2008 is smaller in the simulation than that depicted by the MODIS observations.*

200 **Response:** This is an artefact related to the contouring. Had we used different contour ranges for 2008 and 2018 AOD anomalies, this would have highlighted agreement between MODIS and GEOS for each individual event. The zonal anomaly plots (right-most column) show that for each event, MODIS and GEOS spatial distributions are remarkably similar. We have updated the figure using different contouring for the 2008 and the 2018 events.

– **Comment:** *The model anomalies computed against climatology or 0x emission are very similar in 2018 event but not in 2008. Why?*

205 **Response:** Essentially because the 2018 anomalies are much larger than in 2008, hence they are much more likely to overcome the natural variability in aerosol emissions, i.e., from passive degassing events and changes in meteorology. We have discussed this point in the paper, and it is now further emphasized in the revised manuscript.

– *page9-10: analysis of Figure 3*

210 – **Comment:** *The discrepancies between the simulated anomalies and the MODIS anomalies are larger for cloud fraction than for AOD (Figure 2)*

215 **Response:** Yes, this is expected. The AOD anomalies are primarily a function of the aerosol load and largely determined by the volcanic events. Cloud fraction on the other hand is influenced by many factors including convection, sea surface temperature, ENSO state, cloud microphysics and winds, and it is much more sensitive to natural variability. Satellite retrievals are also influenced by empirical definitions of cloudy/non-cloudy regions, adding uncertainty (Pincus et al., 2012). Given this, it is remarkable that the climatological CF is in

relative good agreement with the satellite retrieval demonstrating the skill of GEOS in reproducing clouds during the volcanic events. This point has been further emphasized in the revised manuscript.

- 220 – **Comment:** *page 10, line 1: I would de-emphasized “reproduced the spatial distribution”: The spatial patterns shown by the simulation are quite different than those shown by MODIS (at least visually, better consistency is shown in the profile).*

Response: We have clarified the text as follows:

... reproduced the spatial distribution of the CF anomaly from the MODIS retrieval (within the plume domain as well as zonal means) ...

- 225 – **Comment:** *page 10, line 5: Why having no correlation between 1x-0x anomaly and retrieval anomalies implies that the observed CF was mainly driven by meteorological variability? Uncertainties in MODIS observations should also be discussed to put into perspective these findings which strongly rely on the accuracy of the observations.*

230 **Response:** Correlation between observations and the 1x-0x anomaly would indicate that volcanic emissions forced ACIs that were decoupled from meteorology - i. e. the observed effects would not have been present without elevated emissions. It is likely that the MODIS CF retrieval has a low bias (although this is in part accounted for by using the satellite simulator) (Pincus et al., 2012), which would move the MODIS anomaly further away from the 1x-0x difference. This is a strong indication that natural variability, instead of the volcanic events, caused the observed anomaly in CF. We have clarified these points in the Section.

- 235 – **Comment:** *Figure 4: please could you indicate the meaning of each figure in the caption, we guess that delta SCF and CF refer to the anomalies?*

Response: Done.

- Section 5.0.1:

- 240 • **Comment:** *This section and the following one are difficult to follow. The back and forth between results and their interpretation makes the reading quite hard. I would suggest commenting first the results and then interpret them in terms of impact on liquid processes.*

Response: We have made an effort to reorganize this section by describing how results are organized in figures/tables in paragraph 1, presenting the results in paragraphs 2-3, discussion in paragraphs 4-5, and separating the presentation of results/discussion for each event where possible.

- 245 • **Comment:** *page 12, line 13-15 “suggesting that ACI for 2018 were not limited ...”: why? additional explanations are needed*

Response: Total water path (TWP) is the sum of liquid water path (LWP) and ice water path (IWP), therefore significant effects in TWP indicate ACIs in both liquid and ice clouds. We have clarified the text as follows: “MODIS TWP anomalies (where TWP is the sum of LWP and IWP) ...”.

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- **Comment:** *page 12: TWP is used here but defined on page 13. The variables, their symbol, unit and meaning should be defined in the data section and in a table.*

Response: All datasets/acronyms have been defined in Section 2: Data. We have also added a looku[table defining each variable.

- **Comment:** *Section 5.0.2: Same remark as for liquid cloud.*

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Response: Thank you for your comment. We have made an effort to rewrite this section to, wherever possible, separate the presentation of results for each event and structure each paragraph such that results are presented first, then discussed.

Conclusion

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- **Comment:** *The second paragraph should be improved.. For example, to understand the line 8-10 on page 28 one should know that SO2 emission was actually 5 times larger in 2018. One should be able to understand the Conclusion without reading the rest of the text.*

Response: Thank you. We have reorganized the conclusions such that overall findings are presented, after which specific findings related to liquid clouds, ice clouds, and microphysical processes are presented separately. We have clarified the text in the example given by the author in the comment above.

10. Technical Remarks

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- **Comment:** *please check the section numbering: a title for section 1 is missing, in section 1 , 1.1.1, 1.1.2 should be replaced by 1.1, 1.2... see also section 4*

Response: Done.

- **Comment:** *I suggest to include a Table giving the meaning of the symbols and acronyms.*

Response: We have added a table defining each variable.

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- **Comment:** *There are a lot of acronyms and symbols. I think that for abstract and conclusions the acronyms should be avoided to facilitate the reading.*

Response: No acronyms were used in the abstract. In the conclusions, we have avoided using acronyms except in cases where they were originally used more than once. In such cases, we define the acronym with the first usage.

- **Comment:** *Overall the quality of Figures is good.*

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Response: Thank you.

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