

1- Introduction

This study aims at characterizing the effects of sulfate and ash aerosol emission on macro and microphysical ice and liquid cloud properties during 2 degassing events from the Kilauea volcano in June 2008 and May 2018. The rationale for analysing and comparing these two events is that sulphate aerosols are mainly originated from the volcanic eruption which allows to separate the impacts of aerosol emission on clouds and climate from other confounding effects such as meteorology and anthropogenic emissions. The methodology relies on the comparison between i) anomalies of simulated cloud properties computed against climatology or no volcanic emission experiment and ii) anomalies of cloud properties derived from MODIS and CALIPSO satellites observations, for both 2008 and 2018 events. A cross-comparison of both events is undertaken to characterize the aerosol cloud interaction signatures in response to volcanic aerosol emission and meteorology. The statistic significance of the results is analysed. The main outcomes of this work are

- The increased in cloud condensation nuclei (first indirect effect) in response to increased aerosol concentration was likely decoupled from local meteorology effects while changes in precipitation efficiency and cloud lifetime (second indirect effect) were limited by meteorological variability.
- For the 2008 event aerosol emissions have affected liquid cloud only while the higher injection height of sulfate and ash for the 2018 event leads to modification of ice cloud.
- The injection of volcanic aerosol triggers deepening of the clouds.
- The GEOS model tends to underestimate shallow cloud and overestimate cirrus clouds
- Importance of accounting for ice nucleation on ash particles to represent cirrus processes

This work contributes to a better understanding of the role of meteorology and microphysics in aerosol-cloud interaction processes which represent an important source of uncertainties in current climate models. Overall, the paper is well written and is within the scope of the ACP journal. The methodology and the data are properly presented. The scientific significance of the results is high enough for publication.

However, I would suggest addressing the following general remarks and specific comments to improve the quality of the paper prior its publication.

Congratulation for this very interesting work.

2- General comments

Could you address the following questions and if needed make the necessary modifications in the text.

- 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.
- 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 ?

The manuscript suffers from lack of detailed information or clarification, please address my specific remarks (next Section) to improve it.

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. subsections 5.0.1 and 5.02).

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.

3- Specific comments

Introduction

- Introduction, line 20 page1 : 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)
- 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 ?
- The outline of the paper should be given at the end of Introduction on page 3 this will give a transition with Section1.

Section 1

- 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.

Section 2

- page 5, line 20: It is not clear what processing is applied to missing values, is it gap-filling ?
- the description of the satellite products given in page 5 is not accurate enough.
 - o 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).
 - o What is the vertical resolution of CALIPSO data ?
 - o What is the temporal frequency of the CALIPSO product ?
 - o How the anomalies (shown in Fig 5) have been computed for CALIPSO
 - o 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 ?
 - o 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)
 - o what are the uncertainties associated with the MODIS and CALIPSO products ? Could you provide product evaluation references ?

Section 3:

- 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.
- page 6, line 9-10: How is volcanic SO₂ constrained by OMI data ? (data assimilation ?)
- 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
- Is the cloud microphysic scheme a GEOS component ? What is the name of the scheme ?
- Is the GEOS model constrained by data assimilation, particularly for aerosol (MODIS AOD ? ...)
- 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

Section 4

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

Section 5

- page 9, line 8-12: The following findings are missing from the analysis of Figure 2
 - o 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.
 - o The model anomalies computed against climatology or 0x emission are very similar in 2018 event but not in 2008. Why ?
- page 9-10: analysis of Figure 3
 - o the discrepancies between the simulated anomalies and the MODIS anomalies are larger for cloud fraction than for AOD (Figure 2)
 - o 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).
 - o 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.
- Figure 4:
 - o please could you indicate the meaning of each figure in the caption, we guess that delta SCF and CF refer to the anomalies ?
- Section 5.0.1 :

- 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.
- page 12, line 13-15 "suggesting that ACI for 2018 were not limited ...": why ? additional explanations are needed
- 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.
- Section 5.0.2: Same remark as for liquid cloud.

Conclusion

- 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.

Technical remarks

- 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
- I suggest to include a Table giving the meaning of the symbols and acronyms.
- There are a lot of acronyms and symbols. I think that for abstract and conclusions the acronyms should be avoided to facilitate the reading.
- Overall the quality of Figures is good.