

Interactive comment on “Systematic satellite observations of the impact of aerosols from passive volcanic degassing on local cloud properties” by S. K. Ebmeier et al.

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I would like to thank both reviewers very much for their detailed, insightful and constructive comments on our manuscript. They have significantly improved our study.

I have addressed reviewers' specific comments individually below:

Reviewer 1

General Comment: This paper builds on an active body of research that aims to understand and quantify the indirect effect of volcanic aerosol on downstream cloud properties. What sets this paper apart from other satellite-based stud-

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ies is the considerably longer observation period, analysis of more active volcanic islands, and attempt to distinguish orography using multiple control islands. While the authors provide consistent results that strengthen conclusions made by previous studies additional steps could be made to forge new discoveries. By incorporating additional data (e.g., CERES and MACC discussed below) more could be stated about the potential implications of volcanic aerosol emissions on present day indirect radiative forcing. This paper could also benefit from improvements to the writing and clarification of some technical details.

I have incorporated CERES data into my analysis, as suggested. I have also clarified several of the technical details, as suggested by both reviewers as described in more detail below.

Major comments:

Throughout the paper it is stated that isolated volcanic islands capture the aerosol indirect effects of the pre-industrial atmosphere. However, it is difficult to determine how important these volcanic islands are on the Earth's radiation budget when explicit indirect forcing estimates have not provided in this paper. Therefore, I suggest adopting the approach described in Yaun et al. (2011) in which they use CERES (Cloud and the Earth's Radiation Energy System) monthly mean observations to derive the top of atmosphere shortwave radiative flux. They find a 20 W/m^2 perturbation caused by the Kilauea plume. Similarly, this approach could be applied to the additional islands examined in this study to determine if the same sensitivity is observed at other locations. It might be beyond the scope of this study but it would be fascinating to examine volcanic plumes near major sources of anthropogenic activity (e.g., along coasts) to demonstrate (by comparing the responses) that, indeed, remote volcanic islands represent the “pre-industrial” atmosphere conditions. These additional steps would certainly make this paper more appealing to the scientific community.

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For each of the volcanoes (and control islands) in our study I have used CERES Single Scanner Footprint (SSF) data to investigate the time averaged Top of Atmosphere Short Wave flux. I used the SSF data due to its higher resolution ($20 \times 20 \text{ km}^2$), rather than the monthly mean observations as used by Yuan et al., 2011. I find a perturbation of $20\text{--}45 \text{ Wm}^{-2}$ (mean = 28 Wm^{-2}) at distances $> 150 \text{ km}$ from Hawai'i and smaller effects at Yasur and Piton de la Reunion. These results are shown in Figure 5 of the amended manuscript and discussed in the Results (3.1-3.3) and Discussion sections (4.3 and 5).

Although I agree that a comparison of volcanic plumes near sources of anthropogenic aerosol to more 'pristine' environments would be very useful, I think it is beyond the scope of this particular study, as the reviewer suggests.

It is mentioned briefly in the manuscript that averages (of cloud and aerosol properties) are constructed over 6-10 years of data (Pg2681 L6-9 and PG 2682 L28). How exactly is the sampling distributed over this time period? How does the sampling vary over the seasonal cycle? As I understand it, both cloud and clear-sky conditions are necessary so that both aerosol and cloud properties can be analyzed within the satellite field of view. How often does this condition fail? My concern is that these types of failures may be weighted more heavily during certain times of the year when metrological conditions produce either too much cloud or not enough in the lee of the island. Finally, the continuity of statistical sampling (over the seasonal cycle) needs to be discussed in greater detail. The sample size could also be biased due to atmospheric vortices caused by the wakes that islands generate. A prominent example of this is the two large quasi-steady counterrotating eddies that extend several hundred kilometers downwind of the big island of Hawaii (Smith and Grubisic, [1993], JAS). The sampling bias for Hawaii is probably small because this feature is quasi-stationary over the annual cycle, however the lower summit heights and substantial variation in thermodynamic conditions at other island locations may cause inconsistent wake

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patterns that could compromise the interpretation of the results in this study.

Thank you, this is a very good point. I have taken several measures to address the impact of seasonal sampling bias to this study. Firstly, I have plotted the number of days with good retrievals according to month of the year (Figure 2 in attachment). I have also divided all of the averaged data according to season (split into 3 month intervals Dec-Feb, Mar-May, June-Aug and Sep-Nov) so that all results are presented in terms of 'seasonal averages'. Figure 3 (MODIS) and 4 (AATSR) of the amended manuscript now show aerosol and cloud properties for each volcano divided according to season (Differences in seasonal upwind and downwind values are now also presented in Table 2). Although there is variability in the absolute values of both cloud and aerosol properties between the seasons in all cases, the general trends that are indicative of a volcanogenic aerosol indirect effect persist. The values for aerosol and cloud properties quoted throughout the manuscript now refer to seasonal averages.

Both changes in meteorology and aerosol concentration exert significant control on the properties of low-clouds. For example, as the lower troposphere stability decreases clouds become more convective thereby causing the effective radius to increase; similarly, a decrease in aerosol optical depth causes effective radius to increase (Lebsock et al. [2008], JGR). Thus, it is essential to examine the effects of meteorology on downstream cloud properties as a way to rule out mechanisms that could be mistakenly interpreted as an aerosol indirect effect. I suggest examining the response as a function of the Froude number. Froude number is useful to diagnose whether the airflow will be forced up over the mountain causing trapped-lee waves or "blocked" and around causing lee-vortices downstream (Etling, [1989], Meteorol. Atmos. Phys.; Schar and Durran, [1997], JAS; Epifanio, 2003, Encyclopedia of the Atmospheric Sciences). The type of generated lee-waves may affect the sampling and properties of the retrieved aerosol and clouds. This should be investigated (as a function of Froude number or some other thermodynamic parameter) to strengthen the argument

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made in the paper that these orographic contributions to the mean cloud properties are indeed smaller than the influence from volcanic aerosol. Froude number can be calculated from ECMWF reanalysis data for the atmosphere below 700 hPa (Hughes et al. [2009], JAS provides a nice description of calculating the Froude number).

I have divided the data according to unsaturated moist Froude number estimated from ECMWF data for Kilauea, Yasur and Piton de la Fournaise. Figure 10 in the amended manuscript shows COD and CER for a range of Froude numbers. Although COD is higher at greater Froude number at Yasur and Piton de la Fournaise, atmospheric stability seems to have a limited effect on systematic differences between CER upwind and downwind. This is now discussed in Section 3.4

The manuscript is rather lengthy and could easily be shortened by 20 per cent without losing any essential content. For instance the authors discuss, several times, methods in the paper that they considered to apply in their research however did not actually carry out. This type of information is extraneous. For example, pg2680 L15-20 discusses adopting a ship track finding algorithm to find volcano tracks but does not use it for the research here. Is this information necessary? Another example is the discussion about the use of trajectory analysis on Pg's 2697 to examine indirect effects near continents. I recommend removing both of these discussions or describing them in a separate paragraph aimed to establish future research initiatives.

I have revised the manuscript to improve the quality of the writing and to avoid repetition. I have removed both of the statements that the reviewer refers to and now discuss future work directions only very briefly in the concluding section of the paper (5).

Minor Comments:

Pg. 2676: L1: I believe you are referring to the research of Schmidt et al. 2012, ACP. If so, you need to specify that the "significant source of uncertainty" comes

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from a global aerosol microphysics model. and **Pg. 2676: L5:** The line "Understanding the impact of volcanic emissions on indirect radiative forcing is important: : :" might be overstating its effect on climate. It is not clear how important it is because few studies have estimated the forcing (all of which used modeling data that is typically fraught with cloud property biases). Instead, the intended meaning could be changed appropriately by replacing the bold faced words is important with provides a tool to study.

The abstract make it simpler and more precise. The first two sentences now read: "The impact of volcanic emissions, especially from passive degassing and minor explosions, is a source of uncertainty in estimations of aerosol indirect effects. Observations of the impact of volcanic aerosol on clouds contribute to our understanding of both present day atmospheric properties and of the pre-industrial baseline necessary to assess aerosol radiative forcing.."

Pg. 2676: L9: Acronyms (MODIS and AATSR) should be spelled out when first defined.

Done.

Pg. 2676: L10-11: It is unclear what it means that the retrievals were rotated in this sentence. Specify that the retrievals were rotated about the volcanic vent to be parallel to the wind direction. Also, it's not clear in this sentence why signal to noise is improved without digging into the manuscript. Wasn't the rotation needed so that the upstream retrievals could be compared to the downstream retrievals?

This has been clarified and now reads: "Retrievals of aerosol and cloud properties at ... are rotated about the volcanic vent to be parallel to wind direction, so that upwind and downwind retrievals could be compared."

Pg. 2676: L19-20: This sentence makes it sound like this is the first study to

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examine the volcanic indirect effect when it is not. This research does however present new information that should be highlighted here instead. What sets this paper apart from Gasso 2008 and Yaun et al. 2011 is that it provides a considerably longer observation period over multiple active and non-active volcanic island sites to estimate volcanic emissions on low-cloud properties.

I have clarified this. "This study presents systematic measurements over several years at multiple active and inactive volcanic islands. Our observations of unpolluted, isolated marine settings may capture processes similar to those in the pre-industrial marine atmosphere."

Pg. 2677: L10-11: Evaporation of small droplets in polluted clouds can also be invigorated if the overlying air is sufficiently dry (Ackerman et al., 2004, Nature).

I have added the following description of cloud lifetime effect: "If the overlying air is sufficiently dry, the evaporation of small droplets in a polluted cloud is enhanced, so that cloud water content decreases as droplet concentrations increase (Ackermann et al., 2004)"

Pg. 2677: L11: Should specify that this is the change in the Earth's radiative balance "as CO₂ levels rise." The aerosol indirect effect is minuscule compared to the incoming and outgoing radiative fluxes at the Earth's surface.

This has been corrected to: "Carslaw et al. (2013) suggest that 45 per cent of variance in post-1750 aerosol forcing (the contribution of aerosols to the change in the Earth's radiative balance as CO₂ levels rise) is from natural sources, which are hard to isolate and measure in the polluted present-day atmosphere (Andreae et al., 2007)."

Pg. 2677: L23-24: I think the intended meaning of this sentence is to suggest that studies examining the "local effects of aerosols on clouds" is dominated by ship track studies. There are of course numerous studies that examine the indirect effect on global scales (e.g., Chilson et al. (1992), Science; Lebsock et

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al. (2008); JGR, Quaas et al. (2008), JGR Bellouin et al. (2013), ACP to name a few).

I have corrected this: "Direct observations of the local effects of aerosols on clouds have so far been dominated by measurements of ship tracks"

Pg. 2677: L27: The statement "the impact of the polluted clouds are otherwise identical in origin and thickness to clean clouds in the surrounding cloud deck" is not always true as pointed out by Christensen and Stephens, 2011, JGR where they observed that the polluted clouds are often elevated in height compared to the unpolluted clouds.

Thank you, this has been corrected as follows "Aerosol from shipping provides an ideal experiment for isolating the impact of aerosols, as the polluted clouds are similar in origin and thickness to clean clouds in the surrounding cloud deck, although they may be elevated in height (Christensen and Stevens, 2011)."

Pg. 2687: L17: Please quantify the words "high proportion" that degassing volcanic SO₂ flux emissions make up in the atmosphere.

The words "high proportion" (30-70 %, according to different studies) refer to the contribution of passive degassing to total volcanic SO₂ flux, not the proportion of volcanic SO₂ to total SO₂ sources: "Time-averaged emissions from passive degassing are thought to make up 30-70 % of the volcanic SO₂ flux to the atmosphere (Andres and Kasgnoc, 1998; Halmer et al., 2002; Mather et al 2003)."

Pg. 2681: L3, L6; L12, L15, L20, and elsewhere; the word 'we' is used repetitively and is distracting - consider adopting other writing styles to enrich the language in the manuscript.

I have rewritten this section and other parts of the manuscript to avoid repetition of ideas and sentence structures

Pg. 2682 L1: Don't you mean cloud top pressure > (greater than) 440 mb?

C5546

Thank you –corrected

Pg. 2682 L2: What do you mean by sampling rate of around 100-300 per bin? Is this the average number of samples you get in a 10 km grid box per day?

This sentence has been removed. I have added charts showing the number of retrievals used in the analysis divided according to month to Figure 2.

Pg. 2682 L8: How often does ECMWF output metrological fields? Are you using the 8-times daily product? Is this the Interim or ERA product? Please specify.

This has been clarified: "Horizontal wind velocity components from the European Centre for Medium-Range Weather Forecasts (ECMWF, ERA-Interim, e.g. Dee et al., 2011) are used to rotate the aerosol..." and "ERA-Interim horizontal wind velocity components (available 5 times per day) are selected for the time of day closest to the satellite overflight."

Pg. 2682: Have you considered using the MACC (Monitoring Atmospheric Composition and Climate) aerosol ECMWF reanalysis product to examine the background composition? MACC is a framework developed by ECMWF Integrated Forecast System (IFS), which fully couples a numerical weather prediction model with data assimilation of satellite aerosol optical depth. The great advantage here is that aerosol optical depth retrievals can be obtained in regions with clouds. Incorporating this dataset would boost the number of samples for your study providing more robust statistics as well as offering an independent measurement of the aerosol indirect effect.

MACC reanalysis provides aerosol optical depths at a horizontal resolution of 80km. This may be sufficient to detect the local increase in AOD downwind of a passively degassing volcano (e.g. at Kilauea and Yasur, where peak AOD are ~100 and 50 km downwind, respectively). However, the MACC aerosol data would be of much lower spatial resolution than the cloud data (MODIS, AASTR) to which we would be compar-

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ing it. I think that a future MACC study could give useful insights into volcanic emissions into the atmosphere, especially as it would allow sulphate aerosol to be identified directly, and as the reviewer suggests, allow an estimation of the Twomey effect from coincident cloud and aerosol measurement. However, I feel that such a study would be best suited to periods of high emission, as the local effects from background emissions examined here would constitute just 1-3 MACC pixels at some of the volcanoes.

Pg. 2684: L22-23: Is the standard error larger for AATSR because the footprint size is larger? Please state the reason why smaller sample size occurs compared to MODIS.

"However, seasonal differences vary between the instruments and AATSR results generally have much higher standard errors than MODIS due to longer repeat time (3 days relative to 1 day), shorter period of coverage (6 relative to 10 years) and consequently smaller sample size."

Pg 2685: L1-5: I applaud the use of multiple independent measurements in the research; it strengthens many of these findings. However, more information needs to be provided about the retrieval. Please specify the wavelengths that are being analyzed for each satellite sensor. For example, MODIS retrieves effective radius at three different wavelengths (i.e., 1.6 μm , 2.1 μm , and 3.7 μm). Which set of wavelengths are you using? I am also surprised to see such large absolute differences between MODIS and AATSR. For example, Chen et al., (2007), JAS, observe only slight variation (1 μm) amongst the MODIS channels referenced above.

I have clarified which wavelengths are being used : "The wavelengths most sensitive to CER are 1.6 μm and 2.1 μm for AATSR ORAC and MODIS (Joint Atmosphere product) retrieval algorithms respectively (Section 3.5). I also mention the discrepancies between the data sets in Section 2: "Some significant differences in CER between these two datasets were observed by Sayer et al. (2011), attributed in part to the different

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wavelengths used by the retrieval algorithms, and consequent differences in sensitivity to cloud vertical structure (e.g. Platnick, 2000)."

Pg. 2686: How does the number of cloud retrievals (upwind and downwind of the volcano) relate to the cloud cover fraction? Have you examined MODIS and AATSR cloud cover fraction variables? Does the volcanic plume increase the liquid phase cloud cover fraction? Fig 4. Indicates that there is considerably more cloud retrievals downstream of the Hawaii volcanic vent, it would be interesting to quantify this for each island. I'm sure the effect is big, Yaun et al. (2008) find a 10 per cent increase.

The cloud retrievals used in our study were selected so as to have cloud fraction >0.2. Figure 4 therefore shows the elevation in number of higher cloud fraction retrievals. I have added Cloud Water Path to the transects of cloud and aerosol properties for each volcano, now Figures 3 and 4. AATSR measurements of liquid water path (and to a lesser extent MODIS) downwind of Kilauea show an increase in liquid water downwind of the volcano. I have added a statement to this effect in Section 4.3: "At Kilauea, and to some extent Piton de la Fournaise, there were an elevated number of cloud retrievals with cloud fraction >0.2 downwind relative to upwind (Figure 4)"

Pg. 2691: L4: define 'a.s.l'

Done - 'above sea level'

Pg. 2692: L4-6. You might find better wind-aerosol relationships using MACC aerosol data product. Separate species (sea salt, sulphate, ect) of aerosol are reported in MACC data. You could also directly estimate the Twomey effect aerosol and cloud properties would be retrieved in the same location.

The aim of investigating the wind-aerosol relationships here was to demonstrate that, in the absence of volcanic emission, sea spray was the most important source of aerosol. I agree that this could be achieved in a more direct manner by using MACC, but I think

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one of the interesting aspects of our study is the change in AOD and cloud properties over tens of metres downwind of the volcanic vents that the higher spatial resolutions of MODIS and AATSR allow us to observe.

Pg. 2694: L18 -28. Please refer to Grandey et al. (2013), ACP for a complete description of aerosol optical depth retrieval artifacts under broken cloudy conditions. Also, these retrieval artifacts will be less affected using MACC data. It thus, may thus be worth considering this dataset for the analysis.

I have added in reference to Grandey et al., 2013: "Artefacts in satellite retrievals of aerosol optical depth under broken cloudy conditions may also lead to spurious correlations between aerosol and cloud properties (Grandey et al., 2013)" Please see comments above regarding the use of MACC data in this study.

Pg. 2696: L8: What is a "volcanic ship track," shouldn't it just be "volcano track"

Corrected, thank you

Pg. 296: L11-12: What do you mean by the words "particular days"? Were some of the days excluded from the analysis? This question leads back to my concern about how the data was sampled temporally and whether all seasons are being sampled equally.

The words "particular days" refer to previous studies than have examined volcano tracks during periods of elevated degassing: Gasso (2008) and Yuan et al., (2011). I have rewritten this sentence to make this clearer: "Our approach builds on previous studies of volcano tracks during periods of elevated activity (Gasso et al., 2008; Yuan et al., 2011) by estimating average volcanic impact over several years"

Table 1: The column "summit height" is somewhat misleading because the emission altitude of the volcanic plume, as in the case of Kilauea, can be significantly lower in elevation than the summit of the island. Can you add the summit height (highest point on the island) and emission height in parenthesis?

C5550

I have added this information into the table

Table 2: Why were the control islands omitted from this table?

These have been added in.

Table 3: What does sigma represent? Is it the standard deviation, standard error, or something else? Also, is this table even needed, these values are already provided in Fig 6, are they not?

Table 3 has been removed

Fig1. I don't understand the caption "using Dark Target and ocean datasets, with Deep Blue to fill in gaps over land?" Are you referring to some particular type of MODIS algorithm or the color scheme used to fill in gaps over the land? Please clarify.

These are MODIS algorithms. The caption now reads: "The multiannual mean (2002-2008) of MODIS Aqua AOD at 550nm, using datasets from the Dark Target and ocean algorithms, with Deep Blue to fill in gaps over land. Grey indicates that there are no data."

Fig2. It is very difficult to read the words on this figure because the characters are very small. If you switch the order of the columns and rows and keep the range on the y-axis the same you would only need to show it once on the left most plot (e.g., show plots b, f, j, n, r, and v in one row; then cloud effective radius in the next row and so on). I cannot read anything inside the picture maps of each island. The valuable piece of information here is the wind rose. I would prefer seeing only the wind rose for each island than being distracted by everything else inside the map. These issues may also become resolved if the figure can be made larger.

I have reorganized this figure, splitting it into one showing maps and now seasonal sampling, and one each for MODIS and AATSR data showing transects for aerosol

C5551

and cloud properties over each volcano and island. This has made the text on the maps larger. I believe that there is valuable information for the reader in the maps, as they give an indication of island size, topographic profile and geometry.

Reviewer 2

General

In this study, the authors investigate aerosol indirect effects (AIEs) resulting from passive degassing of volcanoes located in remote oceanic regions by use of satellite data. The authors do so by employing an analysis technique which allows them to systematically sample aerosol and cloud properties up- and downwind of the emission sources (i.e. the volcanoes). The motivation behind this approach is that mean aerosol and cloud properties downwind of the volcano are expected to be different from those upwind and that these differences are consistent with AIE hypotheses. Because the authors consider volcanoes located in remote oceanic regions, which are assumed pristine with respect to anthropogenic aerosol, this study is a promising step towards characterising the impact of volcanic emissions on properties of the pre-industrial atmosphere. This is especially important in the light of the recent study of Carslaw et al. (2013) who found that uncertainties in quantifying pre-industrial aerosol emissions, and thus the pre-industrial atmospheric reference state, dominate the uncertainty in estimates of total aerosol radiative forcing.

Using their approach of separating polluted from clean environments with respect to volcanic emissions, the authors show that aerosol and cloud properties downwind of passively degassing volcanoes are systematically different from those upwind of the volcanoes. As expected, changes in aerosol properties (an increase of aerosol optical depth, AOD), are more evident than those in cloud properties (reduction of droplet effective radius and increase in optical depth). To substantiate their findings, the authors provide an analysis of "reference

C5552

islands” to exclude the effect of dust emissions and orography on aerosol and cloud properties as well as an elaborate and convincing discussion of the uncertainties of their approach. This paper constitutes an important contribution to the study of AIEs and their quantification from observational datasets, especially as it demonstrates the feasibility of extracting small, but statistically significant signals from long-term satellite data records. The paper therefore fits very well into the scope of ACP.

The paper is generally well written and structured, the motivation and approach are clear and the figures are well chosen and displayed. However, I think the manuscript lacks detail in some instances and some aspects of the results warrant explanation (e.g. not showing results from AATSR for most of the study or the assumptions/conclusions regarding the sampled cloud populations). I recommend the paper for publication in ACP after the following mostly minor issues have been addressed.

P2677, L24-25: Here, I suggest a change/modification to the list of references. Although concerned with quantifying the effect of shipping emissions on aerosol and cloud properties over remote oceans, Peters et al. (2011) did not focus on the analysis of ship tracks. Instead, we aimed at quantifying AIEs from shipping emissions on climatically relevant scales beyond those of individual ship tracks. We did so by systematically sampling for unpolluted and polluted air masses upwind and downwind of major shipping lines. So in a sense, the working hypotheses in the present paper and in Peters et al. (2011) are very similar. However, contrary to the present study, we did not find statistically significant effects of shipping emissions on aerosol and cloud properties in our study. Comparing the two studies, I am certain that the analysis presented here clearly benefits from volcanoes representing a point source, whereas ships obviously represent a moving point source of (in our study) unknown location and strength.

I have removed Peters et al., 2011 from the references relating to ship tracks and
C5553

added the following sentence: “However, Peters et al., (2011) found no statistically significant impact of aerosol from shipping on a large scale, away from the ship tracks themselves.”

P2678, L3-5: I can’t follow your argument in the last part of the sentence.

I have removed this sentence. The comment on challenges in identifying upwind-downwind differences over continents now appears in Section 2.1 (‘Choice of Targets’) in the revised manuscript : “ Uncertainties in AOD retrieved from MODIS data over land are thought to be on average three times greater than over water, where models of surface reflectance are better (Remer et al. , 2005). We therefore focus our study on isolated volcanic islands, avoiding the higher retrieval uncertainties, greater variability in cloud characteristics associated with continents and the systematic dependence of cloud form on wind over coastlines or high topography (e.g. Brenguier et al. , 2003).”

P2681, L3-6: With regards to anthropogenic emissions, the environment of Piton de la Fournaise may not be as pristine as you think. That island is located right along a somewhat major shipping line connecting the southern tip of Africa with Malaysia (see e.g. Peters et al. (2011), Fig. 1). Also, it would be good if you compiled multi-annual maps of cloud cover and cloud top pressure including the associated standard deviations for each of the islands and instruments so that readers get an idea of the sampled low cloud population and variability.

I have corrected the statement originally on P2681 to: “These marine environments include some expected to have very low aerosol burden” and added “La Reunion is located near an important shipping lane (Peters et al., 2011), so it is possible that some anthropogenic sources of sulphate aerosol may be present” to Section 3.3. I have also compiled maps over average cloud top pressure and cloud water path for each island, which will be included in the resubmission as supplementary material.

P2681, L20: Please explain the MODIS QA value of 0. Many readers will not be familiar with it.

I have added in: "...Quality Assurance (QA) values are >0 (removing data where confidence in the retrieval was low)."

P2681, L21: Why did you pick the aerosol product cloud fraction to be <0.8? 0.8 to me seems to be a quite large value and I would assume that at such high cloud fractions, the retrieved AOD could be enhanced due to humidification of aerosol particles in the presence of clouds (e.g. Quaas et al., 2010, and references therein). Did you check if changing the threshold has an effect on the results? For completeness, you may also want to mention the different resolution of aerosol and cloud properties as provided in the ATML2 products (5 x 5 km² for cloud and 10 x 10 km² for aerosol).

I have added in a sentence addressing this: "Although there may be artefacts in AOD retrieved in conditions of broken cloud (e.g. Grandey et al., 2013, Quaas et al., 2010), we use AOD retrievals when cloud fraction is up to 0.8 to maximize the number of retrievals in our analysis" I have added in the ATML2 resolutions to Section 2 ('Satellite Data').

P2682, L1: I wonder if the threshold for cloud top pressures actually has an effect on the results. I would assume that every cloud reaching that high is well above the freezing level and is therefore at least of mixed phase. Those clouds would be already filtered by the cloud top phase criterium, wouldn't they?

True. This does not seem to make a difference to our results. This has been rephrased to " We restrict our observations to liquid water rather than ice clouds (and to data where cloud top pressures were > 440 mb)."

P2682, L2: I am not sure what you mean by 'bin'. Do you mean the 10km resolution the data are resampled to ? If so, it is not clear from the text where those large numbers come from because the data itself has a resolution of either 5 x 5 km² or 10 x 10 km².

C5555

This sentence has been removed. I have added more information about sampling in response to Reviewer 1's comments, including Figure 2 and separation of seasonal average properties in Figure 3 and 4.

P2682, L7: Later in the text, you mention that you use ERA-Interim. This should be noted here as well. Also, please mention the time and spatial resolution of ERA-Interim here.

Done

P2683, L3-4: What do you mean by low sources of uncertainty here ?

I now specify "retrieval uncertainties"

"General features of rotated cloud and aerosol properties": It would be good to mention the results shown in Table 2 at this point. Regarding Table 2, can you also provide corresponding standard deviations? Comparing the numbers between MODIS and AATSR, it seems AOD differences are larger, but CER and COD differences are smaller in AATSR compared to MODIS. Can you comment on this ? Furthermore, the results obtained from AATSR are only really mentioned in this part of the paper, the rest focuses on MODIS(Aqua). Why?

I have added in mention of Table 2 and the beginning of this Section, now called "Data consistency and uncertainty". Table 2 now shows values for both MODIS and AATSR, split according to 3 month 'seasons'. The values in this table are now upwind and downwind differences, and uncertainties are the squareroot of the sum of squares of the standard error of the upwind and downwind values. All results for AATSR data are now shown in Figure 4 (new). The differences between AATSR and MODIS CER absolute values are addressed in Section 3.5, as are the difference in uncertainties.

Section 3 in general: I suggest the volcanoes and their emission profiles be introduced before the actual observed aerosol and cloud properties are discussed.

I have rearranged Section 3, so that the descriptions of results for each of the three

C5556

volcanoes comes before the general description of overall results.

P2685, L27 - P2686, L2: This is also what we found in all the regions we analysed in Peters et al. (2011) and one of the reasons we could not identify statistically significant AIEs from shipping emissions. In light of the plots shown in Fig. 2 of the submitted manuscript, I suggest adding a note to various parts of the manuscript that the observed cloud properties at Yasur do not clearly indicate statistically significant AIEs, but that changes in aerosol properties are evident. This is needed especially because very similar linear trends can be seen for two of the control islands: Fiji and Samoa.

I have added a note of this at several points through the text, e.g at the end of the Section on Yasur (3.2): " Although some of this difference can be attributed to the impact of excess aerosol from Yasur, this is superimposed on a regional trend in aerosol and does not clearly indicate statistically significant aerosol indirect effects. Similarly, the elevation in ToA SW flux downwind of Yasur (Figure 7 c–d) is likely to be influenced by regional variation in cloud properties, rather than purely volcanic effects"; in Section 3.4: "Cloud droplet radius, though influenced by orographic processes, is more strongly affected by volcanogenic aerosol at Kīlauea and Piton de la Fournaise, and regional trends in cloud properties at Yasur"; and in Section 3.5: " This means that although aerosols are elevated downwind of Yasur, there is not evidence of a statistically significant aerosol indirect effect"

P2687, L12: I find it very hard to depict a decrease in droplet size for Tristan da Cunha from the plots in Fig.2.

This statement has been removed

P2688, L3 and many later instances: Sometimes, you refer to emissions from volcanoes in terms of Mg, sometimes in terms of t. For the sake of consistency, could you please stick to similar units throughout the paper?

C5557

I have converted to Mg throughout the paper

P2689, L12-14: The term cloud seeding is normally used for methods which enhance the precipitation efficiency of clouds, therefore reducing cloud cover.

Thank you, this has been corrected : "The growth of cloud droplets associated with volcanic aerosol and the formation of orographic clouds are likely to contribute to this effect."

P2692, L9-10: Why would that be ? Long range transport of especially dust aerosol can occur for 1000's of kilometers under certain conditions. Are there estimates for those kinds of emissions from the considered islands? In any case, I would assume they are low compared to volcanic emissions.

I am not aware of any estimates of these type of emission from the islands considered here. I have removed this statement, but added a reference to other aerosol sources at the end of the first paragraph of Section 4.1 ('Observations of Volcanic Aerosol': " This supports the assumption that sea spray is the most important background aerosol at these volcanoes, rather than, for example, organic carbon or mineral dust from the islands."

P2693, L6-7: An increase in is associated with the Twomey effect. An increase in cloud lifetime would be seen in an increase in liquid water path and/or cloud fraction. It would be informative to show at least one or even both of these cloud properties.

This has been clarified as follows: "Liquid water path derived from retrievals of cloud optical depth and CER reaches its peak value within 50 km of the volcanic vents and is very slightly elevated downwind relative to upwind. This could indicate an increase in cloud lifetime due to drizzle suppression (e.g. Lohmann et al, 2005) but it may also be the consequence of orographic cloud formation downwind or the contribution of cooling and condensing water vapour" (Section 4.3). I have also added liquid water

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path profiles to those shown in Figures 3 and 4.

P2693, L9-13: Can you comment on the influence this might have on the observed cloud properties and why this could be important?

I have added this sentence to the end of the paragraph: "The condensation of evaporated seawater may result in an increase in liquid water content downwind that would mask any evidence of secondary indirect effects." (Section 4.3)

P2694, L6-7: The way you use the data, i.e. level2 products for both aerosol and cloud properties, it is fair to assume that clouds and aerosols are exposed to the same air mass. However, as indicated earlier, this may also mean that AOD is biased high by the presence of clouds. and P2694, L12-14: It is not clear to me what you mean by this. Do you mean cloud cover in general (which I assume would be highly autocorrelated)?

I have simplified this paragraph and removed the misleading statements: "In spite of the strong correlation, our MODIS and AATSR measurements do not allow a direct measurement of the Twomey effect because retrievals of aerosol and cloud properties are mutually exclusive (i.e. aerosol properties are retrieved only where pixels are not flagged as cloud). However, the increase in average downwind ToA SW flux seen in CERES data for all sky conditions shows a radiative impact over a similar area to the downwind cloud and aerosol perturbations, providing additional evidence of a volcanic first indirect effect."

P2695, L7-9: This needs explanation, e.g. secondary sulfate aerosol formed from volcanic emissions of gaseous SO₂ downwind of the volcano is of nucleation or Aitken mode size and thus cannot act as CCN in environments of small supersaturation because that requires at least Accumulation mode sized particles (for typical supersaturations in stratocumulus clouds (e.g. Pierce and Adams (2007))).

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I have added in a sentence of explanation: "The secondary sulphate aerosol formed from ambient reactions of volcanic SO₂ are typically <0.1 µm in diameter, and will therefore be too small to act as CCN where the level of supersaturation is low." (Section 4.2, first paragraph)

P2695, L9-13: I don't agree with this argument. First, you do not show plots of mean cloud cover and cloud top height to substantiate your claim of observing primarily stratocumulus clouds. Second, by filtering your data for liquid water clouds with cloud tops below 440hPa, these clouds may be well above the top of the boundary layer in the regions considered. Thus, you also sample clouds which are exposed to free tropospheric aerosol, and this may very well be of volcanic origin considering that Kilauea and Piton de la Fournaise have summit heights in excess of 1000m. However, these volcanoes also emit from smaller side vents which would definitely be in the boundary layer. Are there estimates of how much of the emitted SO₂ stems from the side vents relative to the main vent? and P2696, L5-6: see above

I have removed the sentence at P2695, L5-6 and the second sentence of P2695, L9-13. This statement now reads: "All of the islands investigated here are in regions where the free atmosphere is dominated by trade winds, except Tristan da Cunha, where westerlies dominate. We expect that the measurements of cloud properties are most commonly from decks of marine stratocumulus over the oceans, with contributions from orographic cloud over land and in the islands' wakes. As our results consist of seasonal and multi-annual averages of retrievals, they contain contributions from days with a range of meteorological conditions. The mean values for COD and CER presented here therefore capture net conditions and may not bear a resemblance to the atmospheric processes on any particular day." I have also compiled a figure showing mean cloud properties over each island for the supplementary material.

Fig. 4: I assume this plot also refers to data obtained from MODIS(Aqua)? The Figure caption has been corrected.

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Technical edits

P2677, L6: air parcel -> parcel of cloudy air

Done

P2677, L8: smaller cloud droplets “may” results in... You should also mention that especially secondary aerosol indirect effect hypotheses are highly debated and far from being verified from observations (e.g. Stevens and Feingold, 2009; Rosenfeld et al., 2014)

I have added a few sentences to this paragraph: “In addition, smaller cloud droplets may result in the suppression of precipitation and therefore longer cloud lifetime, i.e. higher albedo (second indirect effect, Albrecht, 1989), although the importance of this effect is thought to vary between atmospheric regimes (Stevens and Feingold, 2009). The impact of secondary aerosol indirect effects have not been well quantified or verified by observations (Rosenfeld et al., 2014).”

P2677, L14 and anywhere else: indirect effect(s) -> aerosol indirect effect(s)

Corrected, thank you

P2679, L1: significant -> large (significant is a statistical term)

Corrected

P2680, L5: remove the “etc”

Done

P2680, L9: such as “the one published by” Andres and

Done

P2681, L11: What do you mean by “deep”?

Corrected to “aerosol emission into the troposphere”

C5561

P2682, L22-24: Rewrite this sentence

“Days when volcanic activity was elevated according to ground reports cannot be identified from time series of AOD”

P2694, L10: light levels -> solar irradiation levels

Done

P2695, L23: thin -> shallow

Done

Interactive comment on Atmos. Chem. Phys. Discuss., 14, 2675, 2014.

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