Referee #1

We would like to thank Reviewer 1 for his/her comments, which helped us improve the quality of the manuscript, as well as his/her fast response during the discussion phase. We discussed each of the points raised by Reviewer 1 among the coauthors and made the changes in the text accordingly. Below each comment you find our answers and the respective changes made.

1. The title.

I see the WACCM model results provide TOA radiative forcing. However, TOA radiative forcing does not equal to "climate impact". To be accurate and avoid misleading information, I would prefer to use "radiative forcing" instead of "climate impact" in the title.

The title was changed to: 'Stratospheric aerosol layer perturbation caused by the 2019 Raikoke and Ulawun eruptions and their radiative forcing'

2. The IASI based Dso2.

Did you verify your definition of SO2 concertation in a previous study? If yes, please cite it. If no, please verify the definition and comment on the performance before use. We have extended Sect. 2.4, including a new figure, to better explain the definition of our D_{SO2} parameter, which is a new parameter introduced in the present manuscript. Please note that D_{SO2} is not a measurement of the concentration of SO₂ but a very simple "band-difference" to identify IASI pixels where a strong presence of the SO₂ absorption signature can be found. This is in no way a quantitative parameter and is only useful in cases, like the one described here, where a strong SO₂ emission is observed, with the only aim to observe the SO₂ plume dynamics. This is discussed now in the text.

'...R(n) represents the radiance observed from IASI at the wavenumber n. The two values n1= 1129.25 nm and n2= 1130.25 nm represent two spectrally-close wavenumbers, the first at the center of a SO_2 absorption line and the second outside. Figure 1 shows a case of simulated IASI spectra with and without SO₂ (all other parameters in the simulations of the IASI spectra are the same, e.g. surface temperature, temperature and humidity profiles, gaseous absorbers and aerosol profiles). The two selected wavenumbers n1 and n2 are highlighted to show their extreme position (n1 at the approximate center and n2 outside the absorption feature) in one isolated SO₂ absorption line, which is not affected by the absorption of water vapor or other extra-SO₂ species. From the definition of Eq. 1 and Fig. 1 it is possible to see that values of D_{SO2} larger than 1.0 are linked to spectra where SO₂ is detected. It is important to stress that D_{SO2} is purely a qualitative detection parameter is not to be taken as a quantitative retrieval of the SO₂ concentration, even if linked to this latter. This parameter is only useful in case of strong SO₂ anomalies, like the one generated by the Raikoke eruption, and for the analysis of relatively large-scale dispersions of SO₂-rich plumes.'



Figure 1: Simulated IASI spectra with (black) and without (red) SO₂ and a zoom of the SO₂ absorption line used to define the D_{SO2} parameter of Eq. 1.

3. The LOAC data.

Why are the uncertainties explained here different from and worse than the uncertainties in Section 2.3 in Renard et al. (2016)? I assume you used a newer model of LOA

I assume you calculate stratospheric AOD (sAOD) from the LOAC data above the tropopause to 23 km. But you did not make it clear in the manuscript.

We thank the reviewer for their remark which has revealed a mistake in an uncertainty number we provide. The uncertainty is indeed $\pm 20\%$ for concentrations higher than 1 particle.cm⁻³ and not 10 particles.cm⁻³, which is in agreement with the Renard et al. AMT 2016 paper. We have refined the uncertainty values provided in the AMT paper by adding a specific information for submicronic particles; that is why we had written: the uncertainty increases to about $\pm 30\%$ for submicronic particle concentrations higher than 1 particle cm⁻³. We have corrected the mistake in the new version of the text: *'It provides particles number concentrations for 19 sizes in the 0.2 – 50 µm size range, with an uncertainty of \pm 20\% for concentrations higher than 1 particle.cm⁻³'*

Second point: This is true. We changed the Figure caption accordingly 'Derived partial sAODs for balloon borne LOAC aerosol concentration observations from Ury in France, for particle sizes from 0.2-0.7 µm from the tropopause up to 23 km altitude'

4. I would suggest you move section 3, the introduction of the two eruptions, to a more appropriate location, before the CLaMS and WACMM model setting, because it is better to know the date of the eruption, plume height, SO2 volume, etc. before the model set. After moving section 3, please also check the texts and remove the overlapped information of the eruptions in section 3 and in the model setting section. Prior to submitting the manuscript, the authors changed and discussed the position of this section several times. We will move section 3 back before the methods section/ after the introduction.

5. The CLaMS simulation of the dispersion of volcanic plume.

The authors know it very well that the initial plume box for the CLaMS simulation is not accurate, so the simulation results are only suitable for a rough estimation. But this rough assumption would also make the simulation not very necessary. In about half a month after eruptions, the SO₂ concentration and SO₂ plume height would be a nice proxy for volcanic plume dispersion, as you showed in Fig. A3. Or as in https://iasi.aeris-data.fr/so2/. For a longer time after the eruptions, the error of trajectories accumulates and the results are even more unreliable. Are the CLaMS results in Fig.5 supported by the OMPS in Fig. 3 or WACCM simulations in Fig. 4? If not, it would be better to only keep reliable results.

The dispersion of the plume that is simulated with a large number of trajectories follows essentially the evolving analyzed pattern of the atmospheric circulation that is much more reliable than individual trajectories. Numerous previous studies of transport in the lower stratosphere showed that plumes can be predicted one month ahead. It should be also kept in mind that ClaMS is not just a trajectory model but also includes small-scale mixing processes (parameterized depending on the deformation rate in the large-scale flow). Hence, individual trajectories are only calculated over 24 hours (the mixing, or regridding, time step). The reviewer is, of course, right that quantitative comparison between the observations and the simulation without microphysics included is difficult. Our intention when including ClaMS simulation in the paper was to more qualitatively illustrate the pure effect of passive transport on the plume. And the comparison to OMPS indeed shows that passive transport explains the large-scale dispersal of the plume quiet well. However, we weakened the respective statements in the revised manuscript, in order not to overemphasize the comparison between ClaMS and OMPS too much. Relevant sentences are:

'The plume air mass transport is qualitatively largely consistent with OMPS observations, as by the end of July (Fig. 4) enhanced AOD values are apparent throughout all longitudes, mostly north of the Raikoke position. For the CLaMS simulation a clear signal of the tracer is visible around the area of the AMA from end-July until mid-September, which is also consistent with OMPS data (Fig. 4c-e). By mid-August a small percentage of the initialized Raikoke tracer has reached the tropics in the CLaMS simulations...' And the last sentence of this section 'Even though CLaMS simulations neither take any chemical/microphysical processes into account nor possible lifting due to aerosol-radiation-dynamics (suggested to play a crucial role for the Raikoke eruption in Muser et al. (2020)), comparisons show that the horizontal passive tracer distribution from the ClaMS simulation illustrates the effect of passive transport for plume dispersal.'

5. Figure 2

There are very small Dso2 values in the figures, such as in Fig.2a, bottom left corner in Fig.2c, and bottom right corner in Fig.2d. They are probably not SO2 from Raikoke. They may be removed if you only show data with large signal/noise ratios.

As discussed now in the text in Sect. 2.4 and in the reply to major comment 2, D_{SO2} is a purely band-difference detection algorithm, very useful in terms of large-scale analyses of the dispersion of SO₂-rich plumes but not expected to be very accurate at smaller scales. Probably, the small values pointed out by the Referee are false detection due to other spurious spectral signatures (surface emissivity, high clouds, other infrared-radiation-absorbing species or, of course, SO₂ from other sources), which are impossible to filter-out based on retrieval performances. In any case, it is not critical to discuss the first phases of the large-scale dispersion, based on D_{SO2}, which has a clear signature in the Raikoke plume due to the very high SO₂ concentrations in the initial phase.

Specific comments and corrections:

Please make the font of the manuscript uniform.

Please read the manuscript multiple times to correct typos. I list some of them but not all of them here.

Page 1

L1 a moderate stratospheric eruption; *we avoided the term 'moderate now'* L4 short-wave length, high northern ... *ok*

L6 evolution of what? 'has influenced the extent and evolution of the sAOD'

L14 RF. Please spell it out when you use the abbreviation for the first time. *ok* **Page 2**

L21–22 please unify the format of brackets (all half or all full). *Thank you*

L28 Brewer-Dobson circulation

L31 jets

L42 "0.7-2.2" (and many other places in the manuscript): please find out the differences between hyphens and dashes and use them right.

Thank you. This has been done accordingly (not marked in yellow).

L46 Fromm et al. ... This sentence is very confusing, please try to rephrase. *"Fromm et al.* (2014) raise awareness that some limitations in data quality (for the <u>example of OSIRIS satellite measurements</u>), but also conflicting injection sequence information used for potential model studies, which can lead to different conclusions about the same volcanic eruption."

Page 3

L58 setup ok, changed for all cases

L69 "1.5 (Rault and Loughman, 2013)." ok

L71)) Please add reference to MERRA2 data here. ok

L72 Are the OMPS AOD data contaminated by ordinary clouds? Clouds are not filtered for the data set that we use. However, we focus on the stratosphere and therefore clouds do not play a significant role for this study. A sentence has been added for clarification: "To avoid removing enhanced aerosol layers that were mistakenly identified as clouds, we use the unfiltered OMPS dataset. The influence of stratospheric clouds

for the interpretation of this transport study about the Australian fire plume is expected to be negligible and not further analyzed."

L74 L81 dataset

L82 +- --> ± ok

Page 4

L109 "nm" is not a unit for wavenumber. I guess you may want to say "cm–1". Thank vou

Page 4

L85 "marked added-value" Can't understand.

"However, the better vertical resolution and observations on multiple wavelengths compared to OMPS, bring an added-value when spatio-temporally averaged data are used for the radiative forcing calculations."

L95 "The Dust RGB product performs better for volcanic plumes than the Ash RGB product at large viewing angles." Please add a reference here, or explain it if a reference is not available. *Reference added: Eumetrain 2020*

Page 6

L150 "- the" L153 "- The" L154 "- The": I do not understand the usage of

hyphens. This was supposed to represent a sequence. We changed this to numbering. L152 "Mid-latitude" ok

Page 8

L238 moving ok

L239 usually no -> exceptionally

Page 25

Please put Fig. A1 together with other figures in the appendix. It is the ACP Latex template, which places the Figures. So we hope/believe that this will be handled during the formatting step by ACP.

Please put brackets around "10–3" in Fig.A1b to make the X label format uniform. ok And add (km) as Y label for Fig.A1b. ok

The font size of figure titles is not the same. This has been changed for all Figures in the Appendix accordingly.

Page 26

Please add a latitude range for Fig. A2c–d. ok

Please put the acknowledgement together with other texts. Latex ACP template Page 28

Please double check the format of your references ONE BY ONE to make sure they are in the ACP reference format. This is a typesetting issue. More information are given in the original Latex file. Copernicus chooses which information to use for their style.

Page 30

Please avoid citing a paper that you are not sure whether it is finished or not. By now the Khaykin paper is published, the reference is changed accordingly.