Reply to comment by Timothy Garrett

There is a similar phenomenon of green sunsets that is seen for the case of storm clouds as discussed by Bohren and Fraser (1993) https://journals.ametsoc.org/view/journals/bams/74/11/1520-0477_1993_074_2185_gt_2_0_co_2.xml. Specifically, Bohren argues that the color of such sunsets owes to a blue-shift of red sunset light due to a combination of scattering and a preferential absorption by condensed water in the red. Can this explanation be excluded for the case described here? A substantial amount of condensed water would be required, but of course Krakatoa was an exceptional event.

Reply: This is an interesting suggestion, thank you! We considered this hypothesis that the green colours could be caused by liquid water or ice in clouds carefully and concluded that it cannot provide an explanation for the green sunsets after the Eruption of Krakatoa for the following reasons:

1. ***The tropospheric H2O mass is about 13 Tt (Tera tonnes) or 1.3 x 10^7 Mt. The amount of H2O injected by Krakatoa into the atmosphere is not known, but the recent Hunga-Tonga Hunga Ha'apai (HTHH) eruption in January 2022 injected about 150 Mt H2O into the stratosphere. HTHH and Krakatoa are difficult to compare (despite some similarities), but considering the numbers mentioned in the previous sentence, it seems likely that the Krakatoa eruption led to a very minor increase in the amount of H2O in the entire atmosphere. In addition, under normal conditions tropospheric H2O is replaced 40 times per year, i.e. it seems very unlikely that the H2O emitted by Krakatoa led to green sky colours several months after the eruption.***
2. ***The hypotheses by Bohren and Fraser require really large optical depths (and liquid water paths), as mentioned several times in Bohren and Fraser (1993). With such thick clouds twilight phenomena would not occur. This corresponds to an observation that can be frequently made: under overcast conditions, coloured sunsets do not occur.***
3. ***The hypotheses by Bohren and Fraser provide explanations for green thunderstorms only for scenarios where the sun is above the horizon, while the remarkable reports of green colours mainly occur for twilight conditions, i.e. the sun is already below the horizon.***

But it is interesting that your summary, i.e. "a blue-shift of red sunset light" still hits the mark. In case of the green volcanic sunsets, the solar radiation reaching the aerosol layer has been reddened due to Rayleigh extinction and is then "anomalously" scattered by aerosol particles of a suitable size distribution, i.e. scattering by aerosols leads to a blue shift in the spectrum, different from the hypothesis by Fraser and Bohren in Bohren and Fraser (1993).

We added a paragraph in the section 4 (discussion) to discuss the relationship of green sunsets and green thunderstorm clouds.

Reply to review by Filip Vanhellemont

The authors provide an explanation for the observed green twilight observations that were reported by eye witnesses after the eruption climax of Krakatoa in 1883. They use the well-known SCIATRAN radiative transfer code to simulate radiances, based on an educated assumption on the aerosol density profile shape. The perceived color is then estimated by converting radiances to chromaticity values using the CIE color matching functions. The color is studied as function of particle size distribution parameters, total ozone column and aerosol optical depth, for a number of solar zenith angles representative of twilight. The authors arrive at the clear conclusion that green twilight can be simulated for sufficiently large aerosols from a narrow size distribution and sufficiently large optical depth.

General Comments

This article is perfectly suited for publication in ACP. I am not aware of any other publication that presents an explanation of volcanically related green twilight, so the obtained conclusions are important. Furthermore, while it is difficult to obtain precise numbers on e.g. particle size from observations that are subjective (visual color perception), clear constraints/thresholds are obtained on particle size and optical depth. These are new numerical results on an important past volcanic event, and should be published.

> ***Reply: We thank the reviewer for his positive evaluation of our manuscript!***

The applied methodology is clearly explained, and it should be possible for other researchers to reproduce the results. The article title and the abstract perfectly describe the content. The body text is very well written and needs almost no adaptation. Plenty of references are provided.

I really don't see any major problem with this paper; I have a few minor remarks that should be taken into account (see below), so I strongly recommend the publication of this paper.

> ***Reply: Thank you!***

Specific comments

Comment: It is not clear to me at which wavelength Aerosol Optical Depth (AOD) is evaluated in the entire paper. This should be specified.

> ***Reply: Thanks, the wavelength should of course be mentioned. It is now mentioned in Table 1.***

Comment: The finding that green twilight is associated with narrow size distributions suggests that these green colors are only observed in the early stages of the stratospheric aerosol evolution (say, a few months after the event of August 27). Coagulation of particles shifts the distribution to higher size values, but also tends to widen the distribution, but this process takes quite some time. It might explain why no green twilight is reported at later times after the eruption (as far as I can tell from the paper). Perhaps you can add a small comment if you agree with this.

> ***Reply: Thanks for this suggestion. We are, however, not sure how the particle size distribution (PSD) will exactly evolve after an eruption. In another one of our recent papers (Wrana et al., 2023; reference provided below) we investigated the evolution of the PSD after the eruptions of Ambae (2018), Ulawun and Raikoke (2019) and La Soufriere (2021). In three of theses cases, the median (and effective) radius of the stratospheric aerosols – as retrieved from SAGE III/ISS solar occultation measurements – decreased rather than increased as a consequence of the eruption. In addition, the reduction in size seen in the observations lasted for about half a year after the eruption of Ulawun. The accompanying model simulation also reproduced the initial decrease in size (effective radius), but after August 2019 the modelled sizes increased again, probably due to coagulation. Also because of the discrepancy between***

observation and model we are uncertain what role coagulation actually plays and how effective and fast it is to change the PSD significantly. For the Raikoke eruption (2019) an increase in particle size is seen in both observations and model simulations. In summary, we do not really know what exactly could have happened after the Krakatoa 1883 eruption and decided not to mention the hypothesis proposed by the reviewer. But it is certainly a possible interpretation or explanation.

Reference:

Wrana, F., Niemeier, U., Thomason, L. W., Wallis, S., and von Savigny, C.: Stratospheric aerosol size reduction after volcanic eruptions, Atmos. Chem. Phys., 23, 9725–9743, doi.org/10.5194/acp-23-9725-2023, 2023.

Comment: In Fig. 3, the phenomenon of purple light that is often observed after large volcanic eruptions is also simulated, but it is not mentioned in the paper. A small description (one sentence or so) of this finding would enhance the credibility of the method even more.

Reply: Another good point, thanks for pointing this out. The purple colours are now mentioned in section 3.

Technical Corrections

Comment: Line 3: volcanic activity of Krakatau started already several months before August 27. It is perhaps better to speak about the eruption climax of August 27, or something similar.

Reply: Good point. We decided to remove the exact date from the abstract, because otherwise the sentence would suggest that the green sunsets were observed immediately after the eruption. Now only the year of the eruption in mentioned.

Comment: Line 70: This sentence seems to be incorrect. Suggestion: 'Please note that …, while the displayed …'.

Reply: Changed.

Comment: Figure 3: the caption seems to be wrong. Panel (b) shows Total Ozone Column, Panel (c) Aerosol Optical Depth, while the caption indicates the reverse.

Reply: Thanks for catching this – corrected.

Reply to comments by anonymous reviewer #2

Comment: The paper describes a small study which explains why there can be green sunsets after volcanic eruptions. Radiative transfer model simulations are performed for various different aerosol assumptions to explain the green color and to also constrain stratospheric aerosol parameters, i.e. size distribution and aerosol optical thickness. The detailed discussion shows that the found parameters are reasonable. The authors suggest that the method could be used to constrain aerosol properties of historic eruptions, however I suppose that this would be difficult because there are not many reports about the sky colors available and of course also no quantitative observations.

The paper is clearly presented and since it explains the phenomenon of green sunsets to my knowledge for the first time I recommend to publish the paper after very minor corrections (see below).

Reply: We thank the reviewer for his/her positive assessment of our manuscript!

Comment: Often, the term "anomalous scattering" is used. I think that this is misleading because the dependece of the scattering coefficient on wavelength is just a result of well-known Mie theory and not "anomalous".

Reply: We agree that "anomalous scattering" may be misleading, but this is a standard technical term for the case, where the scattering cross section increases with increasing wavelength. It has been used in the past (e.g. Porch et al., 1989) and was not introduced by us. For this reason we would like to keep the term. However, in order consider the reviewer's comment and to clarify this point, we added one sentence to the introduction stating that Mie theory can also be used to simulate anomalous scattering.

Reference:

Porch, W. M., Blue moons and large fires, Appl. Opt., 28, 10, 1989.

Comment: It is not mentioned how the optical properties of the aerosols were computed. I assume Mie theory?

Reply: The reviewer is right, this was not mentioned. Instead, we did refer to earlier papers based on a similar approach. But it is certainly an important piece of information and is now mentioned in section 2.

Comment: The term "volcanic sunset" in the title sounds strange. To me "Explaining the green sunsets after the 1883 volcanic eruption of Krakatoa" sounds better.

Reply: OK, changed.