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 the 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 total tropospheric H2O mass is about 13 Tt (Tera tonnes) or 1.3 x 10^7 Mt. The mass of H2O injected by Krakatoa into the atmosphere is not known, but it may be estimated based on available estimates of the emitted mass of S and typical relationships between H2O and S in volcanic emissions. Some studies suggest that about 15 Tg of S were emitted by Krakatao in 1883, which corresponds to about 30 Tg of SO2. If we conservatively assume a H2O/SO2 mass ratio of 50, the volcanic H2O emissions would correspond to 1500 Tg or 1500 Mt of H2O, i.e. about 0.01 % of the troposphere's H2O mass. Tropospheric H2O is replaced about 40 times per year, i.e. it seems very unlikely that the H2O emitted by Krakatoa led to a significant increase in the occurrence of green thunderstorms several months after the eruption.***
2. ***The hypotheses by Bohren and Fraser require really large cloud optical depths, 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.