

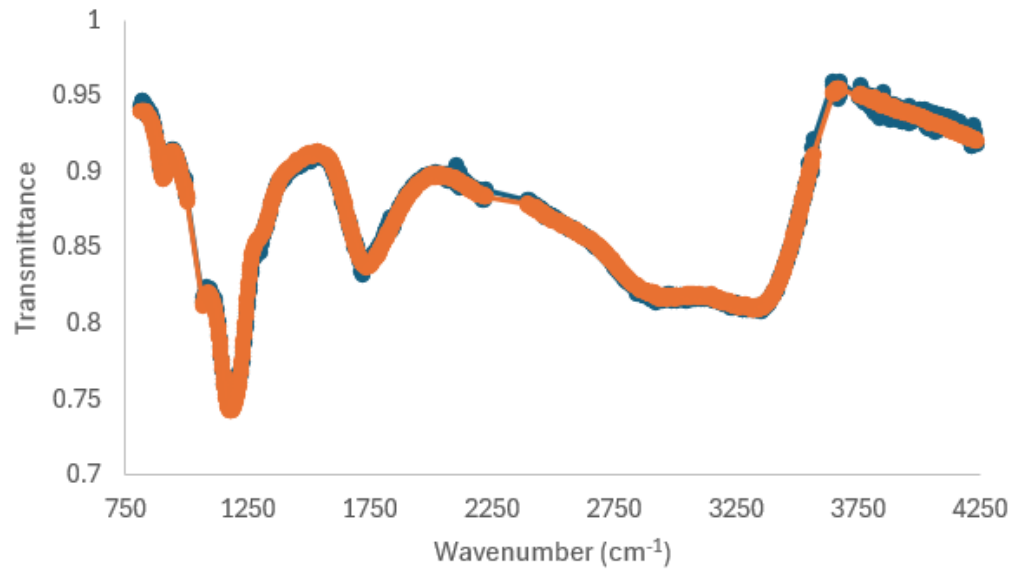
This article describes observations of the Hunga Tonga volcanic plume using high resolution imager measurements from a geostationary satellite. In general, this is a clever approach for extracting atmospheric aerosol information, a bonus set of information that was presumably not targeted in the original mission design. However, there are a few aspects of the interpretation that I don't entirely agree with.

The signals are referred to as reflectances. I expect this comes from standard usage of the data products for Earth observation. The measured signals actually come from scattering by the atmospheric aerosols, rather than reflection, along the lines of limb scattering missions such as OSIRIS. The abstract briefly mentions "scattered light," but for me the terminology used detracted from providing a clear understanding of the nature of the signal. I felt it would have been better to stress that we are dealing with a scattering signal that is quite separate from the normal usage of the satellite's measurements.

If one wanted to generate a quantitative analysis of the signals, I expect the most likely starting point would be SASKTRAN, the freely available analysis software from the OSIRIS team that is geared toward a limb scattering geometry.

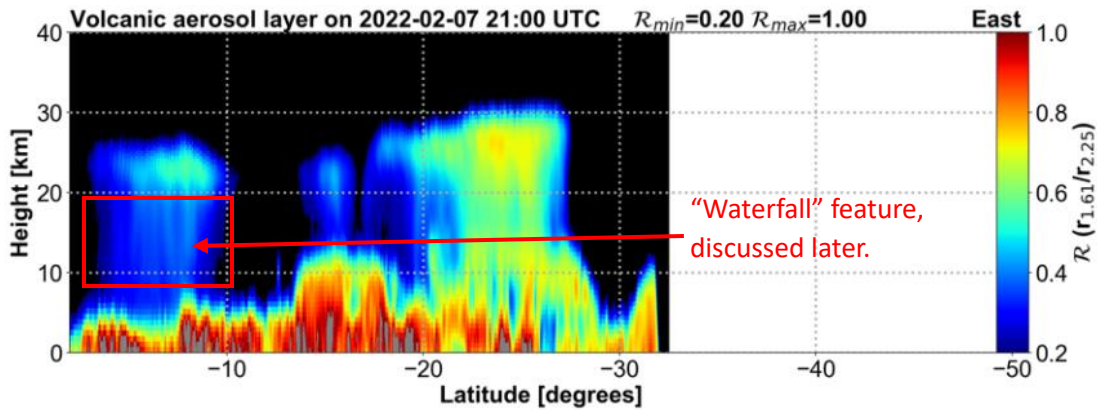
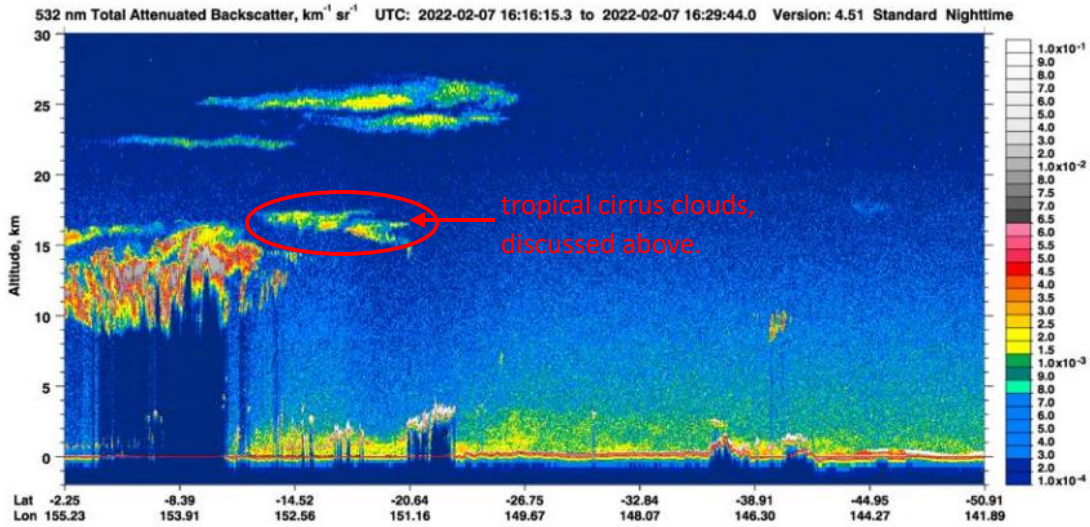
The nature of the aerosols is treated as a mystery (referred to as containing a strong liquid water content), but it really isn't. Volcanic eruptions are well known to create sulfate aerosols, liquid droplet mixtures of H_2SO_4 and H_2O . These aerosols contain a fraction of water, but the presence of sulfuric acid dissolved in the droplet changes the spectral response compared to pure water. The optical constants (real and imaginary components of the refractive index) for sulfate aerosols (aqueous H_2SO_4) are known. They have been measured in the laboratory. It is not appropriate to use optical constants for pure H_2O as a gauge unless one expects liquid water droplets are generating the signal, which is not the case.

I have measured aerosols from the Tonga plume from the Atmospheric Chemistry Experiment. The figure below shows the measured (in blue) and fitted (in orange) results for aerosols observed a few weeks after the eruption [occultation ss99623, measured at latitude 16 S and longitude 166 W on February 7th, 2022, at an altitude of 21.6 km]. The fitted results employ a set of sulfate aerosol optical constants [Lund Myhre CE, Christensen DH, Nicolaisen FM, and Nielsen CJ, Spectroscopic study of aqueous H_2SO_4 at different temperatures and compositions: variations in dissociation and optical properties. *J Phys Chem A* 2003;107:1979–1991, <https://doi.org/10.1021/jp026576n>]. The fact that the measurements can be reproduced accurately using optical constants for sulfate aerosols verifies the aerosol type as sulfate. Over the years that Tonga aerosols have persisted in the atmosphere, there has been some variation in the relative amount of H_2SO_4 and H_2O in the droplets (driven by changes in temperature and ambient water vapor levels), but the aerosol type has remained unequivocally sulfate. You should use refractive index information for that aerosol type when evaluating the spectral response, not refractive index information from a different particle type (like pure liquid H_2O droplets).



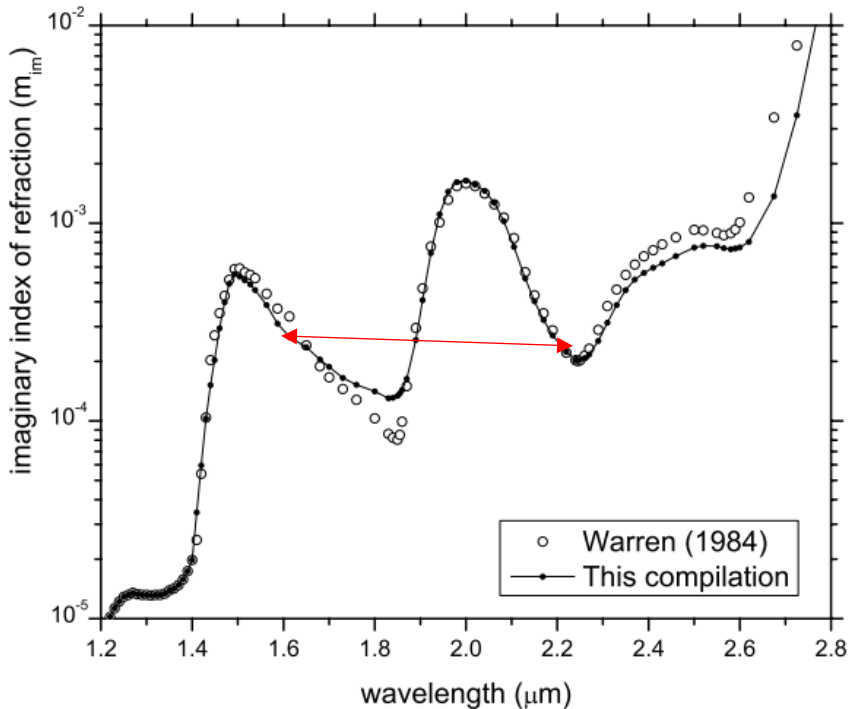
Above: observed and fitted results for ss99623 21.6 km, showing that the predominant stratospheric aerosol type following the Tonga eruption is sulfate.

As for ice, there is some evidence that the approach, using the selected wavelengths, might be somewhat blind to the presence of large ice particles. Below, I have reproduced Figure 7 from the paper, showing the comparison of CALIOP observations with the ratio of signals at 1.61 and 2.25 microns. Circled in red is a feature (around 16-17 km in the tropics) in the CALIOP observations that likely relates to high cirrus clouds, thin clouds composed of relatively large ice particles, a common occurrence in the tropical upper troposphere. As mentioned in the paper, there does not appear to be corresponding signals for any of the tropical clouds in the 1.61/2.25 ratio. While there is a time difference between the two data sets, which means the clouds might have dissipated, this could also point to a similar spectral response at the two wavelengths for ice particles, which would not generate a feature in the 1.61/2.25 ratio.



The figure below is reproduced from the Warren and Brandt paper describing optical constants for ice [Warren, S. G. and Brandt, R. E.: Optical constants of ice from the ultraviolet to the microwave: A revised compilation, *Journal of Geophysical Research: Atmospheres*, 113, 2008]. The arrow indicates the values for the two wavelengths employed in the ratio. They are quite similar, suggesting there could be a low contrast in the spectral response for ice at those wavelengths. A greater contrast could be achieved for ice by bringing in one of the other available wavelengths, such as 0.86 microns, but at the expense of dealing with larger scattering efficiencies, which might complicate the analysis.

WARREN AND BRANDT: OPTICAL CONSTANTS OF ICE



In the reproduction of Figure 7 (2 figures up in this document), comparing CALIOP and the 1.61/2.25 ratio, the box added to the lower portion of the figure highlights a “waterfall” artifact in the results that arises from difficulties in separating out altitude information from the measurements (the reason the results look so diffuse relative to the sharply defined features from CALIOP). This will impact the altitude plots presented in Figure 10, yielding artificially inflated values for the ratio at lower altitudes. Although certainly beyond the scope of this paper, the geometry of the measurement could potentially lend itself to a tomographic analysis [Bourassa, A. E., Zawada, D. J., Rieger, L. A., Warnock, T. W., Toohey, M., & Degenstein, D. A. (2023). Tomographic retrievals of Hunga Tonga-Hunga Ha'apai volcanic aerosol. *Geophysical Research Letters*, 50, 2022GL101978 <https://doi.org/10.1029/2022GL101978>]. If different pixels on the imager provide measurements through the same plume at different angles, supplemented by different views through the plume as the Earth rotates below the geostationary satellite, you might be able to sharpen the altitude discrimination.

In summary, I think this dataset provides a great opportunity for a new aerosol product, if tools such as SASKTRAN were applied to analyze the limb scattering measurements. I felt the nature of the measurements (limb scattering rather than reflection) should have been emphasized more and promoted as the potential source of a new data product. I disagree with the approach of using the wrong optical constants for evaluating the spectral response, discussing ‘high water content’ in terms of refractive index information for pure H₂O, when the aerosols are known to be sulfate, which has different (and known) refractive index information. For ice, I would suggest looking at the ratio of the signals at 0.86 and 2.25 microns to see if a feature appears in the plots from the presumed cirrus clouds. This would verify whether the 1.61/2.25 ratio might have a “blind spot” for ice.