Clouds and Aerosols from the Atmospheric Chemistry Experiment (ACE) and SAGE-III-ISS: Overview and latest results

Peter Bernath, Keith LaBelle, Ryan Johnson

Old Dominion University, Norfolk, VA

and

Chris Boone, Mike Lecours, Jeff Crouse

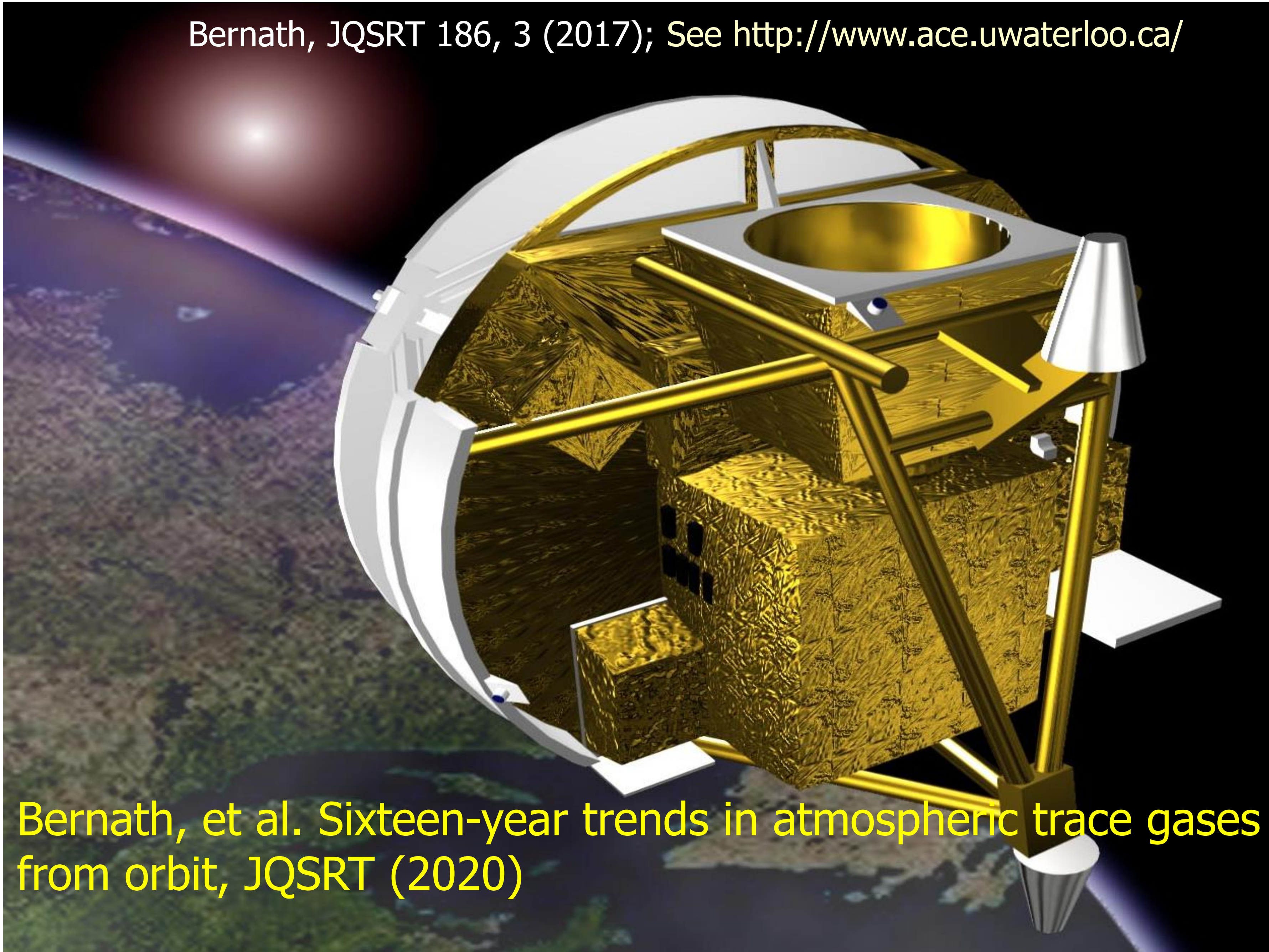
University of Waterloo, Waterloo, ON

SAGE-III/ISS Science Team Proposal

Analyze atmospheric extinction measurements by SAGE-III/ISS and infrared spectra by the ACE-FTS (Atmospheric Chemistry Experiment Fourier Transform Spectrometer) of aerosols with a particular focus on the composition and physical properties of stratospheric sulfate aerosols

Both instruments use solar occultation and are complementary (IR and visible).

ACE Satellite



Bernath, JQSRT 186, 3 (2017); See http://www.ace.uwaterloo.ca/

Bernath, et al. Sixteen-year trends in atmospheric trace gases from orbit, JQSRT (2020)

ACE-FTS Version 4.1/4.2 & 5.0 Species

Tracers: H_2O, O_3, N_2O, NO, NO_2, HNO_3, N_2O_5, H_2O_2, HO_2NO_2, O_2, N_2, SO_2

Halogen-containing gases: HCl, HF, ClO, $ClONO_2$, CFC-11, CFC-12, CFC-113, COF_2, $COCl_2$, COFCl, CF_4, SF_6, CH_3Cl, CCl_4, HCFC-22, HCFC-141b, HCFC-142b, HFC-134a, HFC-23, HOCl, HFC-32

HOCl is a key new ACE molecule for stratospheric ozone depletion.

Carbon-containing gases: CO, CH_4, CH_3OH, H_2CO, HCOOH, C_2H_2, C_2H_6, OCS, HCN, $CH_3C(O)CH_3$, CH_3CN, PAN, high and low altitude CO_2 as well as pressure and temperature from CO_2 lines

Isotopologues: $H_2{}^{18}O$, $H_2{}^{17}O$, HDO, $O^{13}CO$, $OC^{18}O$, $OC^{17}O$, $O^{13}C^{18}O$, ${}^{18}OO_2$, $O^{18}OO$, $O^{17}OO$, $OO^{17}O$, $N^{15}NO$, ${}^{15}NNO$, $N_2{}^{18}O$, $N_2{}^{17}O$, ${}^{13}CO$, $C^{18}O$, $C^{17}O$, ${}^{13}CH_4$, CH_3D, $OC^{34}S$, $O^{13}CS$, ${}^{15}NO_2$, $H^{15}NO_3$

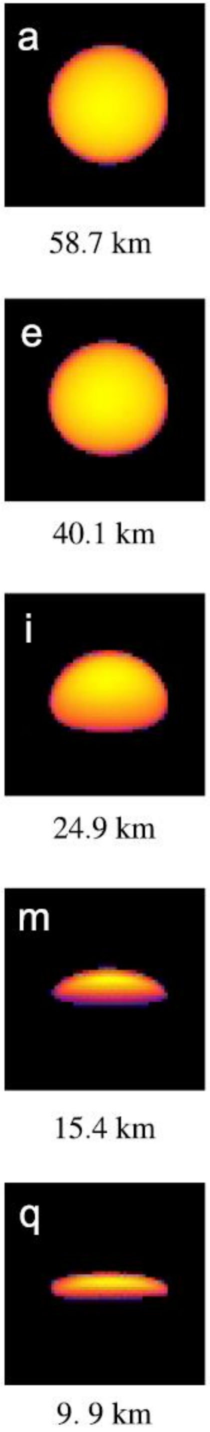
New routine species in v.5.0 are in red.

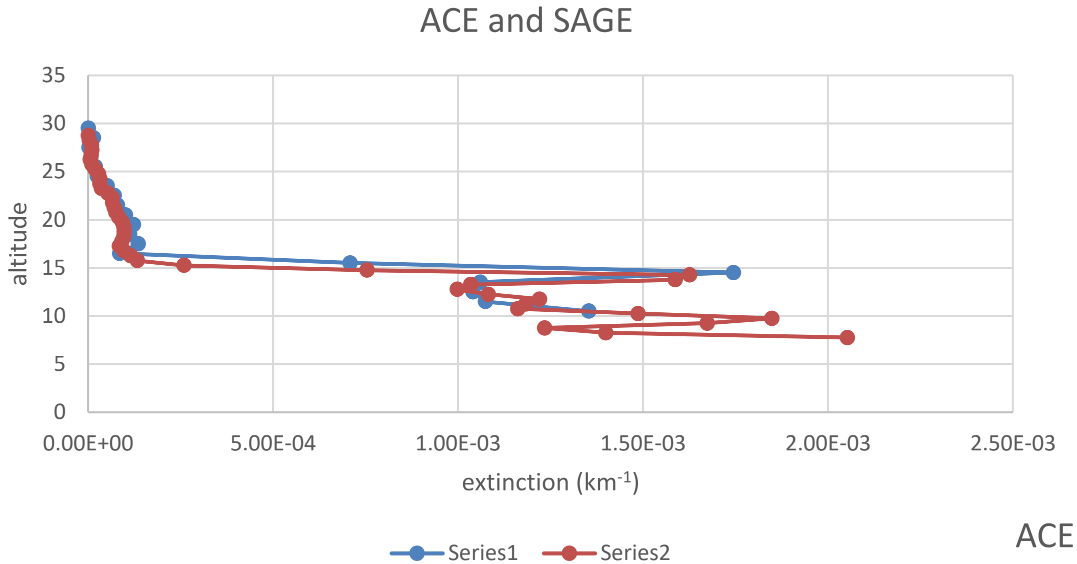
ACE is now in its 19^{th} year on orbit. This longevity makes trend analysis feasible (with care). The change in atmospheric composition is the primary driver of climate change.

ACE retrieves 46 molecules plus 24 isotopologues.

ACE Imagers (1.02 & 0.525 µm)

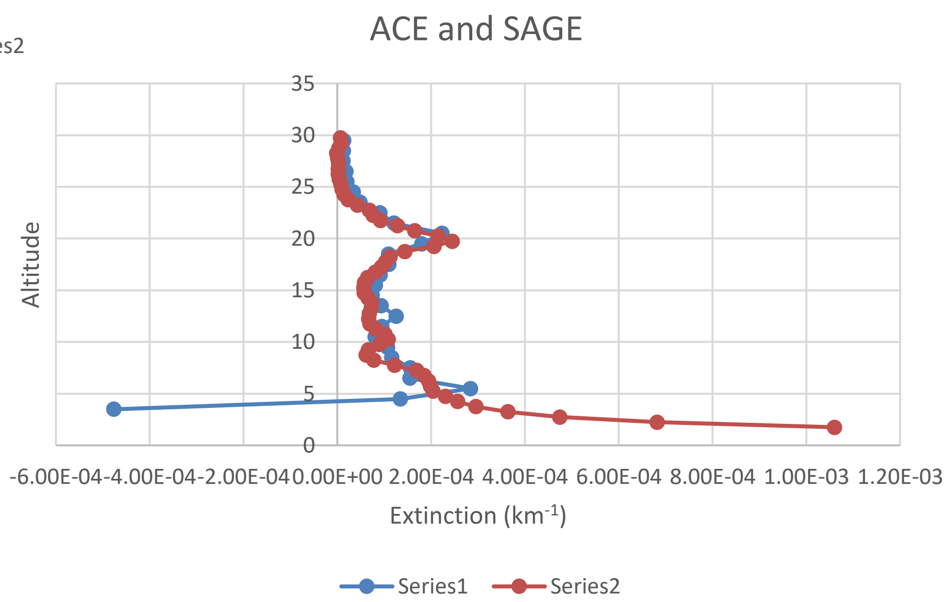
New v. 5.0 of ACE Imager processing provides extinction profiles at 1.02 µm that agree with SAGE-III-ISS values.





ACE (blue)

SAGE (red)



Effects of Smoke in the Stratosphere

Wildfire smoke destroys stratospheric ozone

Peter Bernath[1,2,3]*, Chris Boone[2], Jeff Crouse[2]

Large wildfires inject smoke and biomass-burning products into the mid-latitude stratosphere, where they destroy ozone, which protects us from ultraviolet radiation. The infrared spectrometer on the Atmospheric Chemistry Experiment satellite measured the spectra of smoke particles from the "Black Summer" fires in Australia in late 2019 and early 2020, revealing that they contain oxygenated organic functional groups and water adsorption on the surfaces. These injected smoke particles have produced unexpected and extreme perturbations in stratospheric gases beyond any seen in the previous 15 years of measurements, including increases in formaldehyde, chlorine nitrate, chlorine monoxide, and hypochlorous acid and decreases in ozone, nitrogen dioxide, and hydrochloric acid. These perturbations in stratospheric composition have the potential to affect ozone chemistry in unexpected ways.

Bernath *et al.*, *Science* **375**, 1292–1295 (2022)   18 March 2022

Eight Aerosol and Cloud Spectra

Atlas of ACE spectra of clouds and aerosols

Michael J. Lecours[a], Peter F Bernath[b,a,c], Jason J Sorensen[b,*], Chris D Boone[a], Ryan M Johnson[c], Keith LaBelle[c]

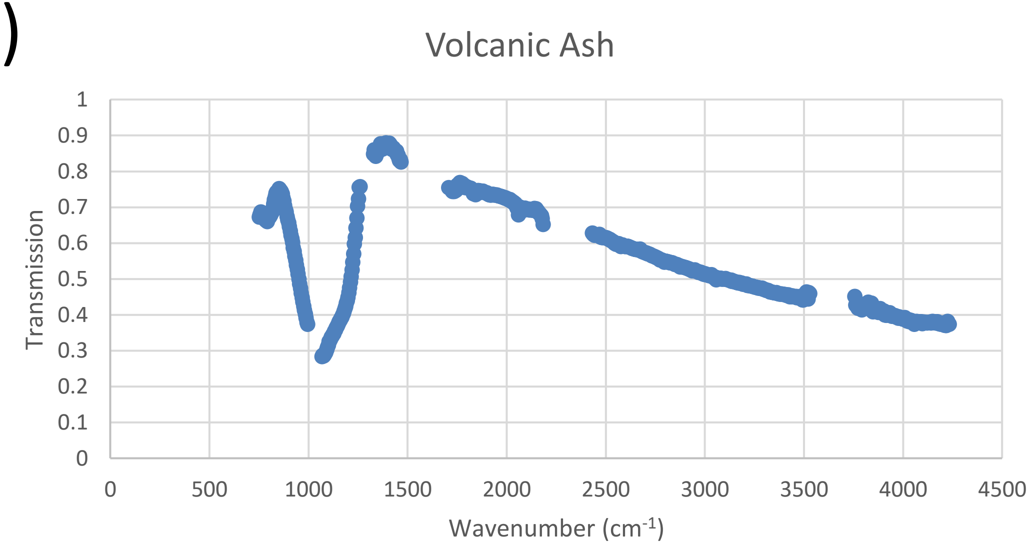
Journal of Quantitative Spectroscopy & Radiative Transfer 292 (2022) 108361

ACE-FTS records characteristic IR absorption spectra of particles

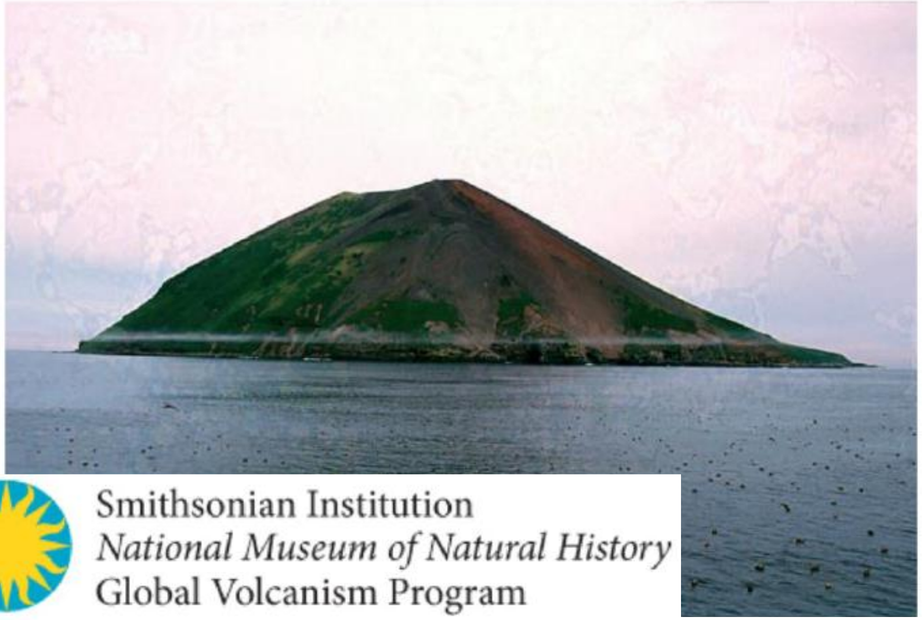
1. Polar Mesospheric Clouds (PMCs)
2. Smoke from fires

Polar Stratospheric Clouds (PSCs)

3. Nitric acid trihydrate (NAT)
4. Sulfuric/nitric acid (STS)
5. Ice
6. Cirrus clouds (ice)
7. Volcanic ash ------------>
8. Sulfate aerosols



Raikoke



Smithsonian Institution
National Museum of Natural History
Global Volcanism Program

Country	Russia
Volcanic Region	Kuril Islands
Primary Volcano Type	Stratovolcano
Last Known Eruption	2019 CE

Latitude	48.292°N
Longitude	153.25°E
Summit Elevation	551 m 1808 ft
Volcano Number	290250

19 June-25 June 2019

Cite this Report

A powerful eruption at Raikoke that began on 22 June (after 95 years of dormancy) was identified based on satellite observations, prompting KVERT and SVERT to raise the Aviation Color Code to Red. A series of at least nine explosions (six within the first 25 minutes) beginning at 0505 and continuing to about 1900 produced ash plumes, with a significant sulfur dioxide component, that rose 10-13 km (32,800-42,700 ft) a.s.l. and drifted E and NE. Lightning was detected in the eruption plumes. Strong explosions at 1640 on 22 June generated ash plumes that rose to 10-11 km (32,800-36,100 ft) a.s.l. The ash and gas was entrained by jet streams and by a cyclone around the Komandorskiye Islands, causing parts of the material to spiral counterclockwise as it drifted farther NE. By 23 June the leading edge of the plume had drifted 2,000 km ENE. According to a news article, at least 40 flights in that region were diverted.

On 23 June ash plumes continued to be visible, rising to 4.5 km and drifting NE. The Aviation Color Code was lowered to Orange. Gas-and-steam plumes possibly with some ash rose to 4.5 km (14,800 ft) a.s.l. and drifted 60 km NW. That same day observers on a passing ship approached the island from the W side; they photographed the island and sent out a drone. An expedition member noted that the entire island was mantled with light-colored ash deposits up to several dozen centimeters thick. In some of the drainages and at the base of some drainages deposits were several meters thick. In some areas along the shoreline waves interacted with the deposits, causing steam explosions and dark brown steam emissions. Gas-and-ash plumes rose 1.5 km above the summit crater rim and drifted W. Minor ashfall was reported in Severo-Kurilsk (340 km NE) during 1830-1920. On 25 June ash plumes continued to be produced, rising as high as 2 km (6,600 ft) a.s.l. and drifting NW.

Sources: Kamchatkan Volcanic Eruption Response Team (KVERT); Sakhalin Volcanic Eruption Response Team (SVERT); NHK (Japan Broadcasting Corporation)

Stratospheric Aerosol Composition Observed by the Atmospheric Chemistry Experiment Following the 2019 Raikoke Eruption

Chris D. Boone[1], **Peter F. Bernath**[1,2,3], **Keith Labelle**[3], **and Jeff Crouse**[1]

[1]Department of Chemistry, University of Waterloo, Waterloo, ON, Canada, [2]Department of Chemistry and Biochemistry, Old Dominion University, Norfolk, VA, USA, [3]Department of Physics, Old Dominion University, Norfolk, VA, USA

Abstract Infrared aerosol spectra derived from Atmospheric Chemistry Experiment measurements following the June 2019 Raikoke volcanic eruption are used to evaluate the composition of stratospheric aerosols in the Arctic. A blanket of aerosols, spanning an altitude range from the tropopause (8–11 km) to 20 km, persisted in the stratosphere over northern latitudes for many months. The aerosols within this blanket were almost exclusively sulfates. The percentage of sulfuric acid in the aerosols decreased over time, dropping below 50% H_2SO_4 concentration at some altitudes by March 2020. Contrary to previous reports, the aerosol blanket was not comprised of smoke particles.

[JGR: Atmospheres: https://doi.org/10.1029/2022JD036600](https://doi.org/10.1029/2022JD036600)

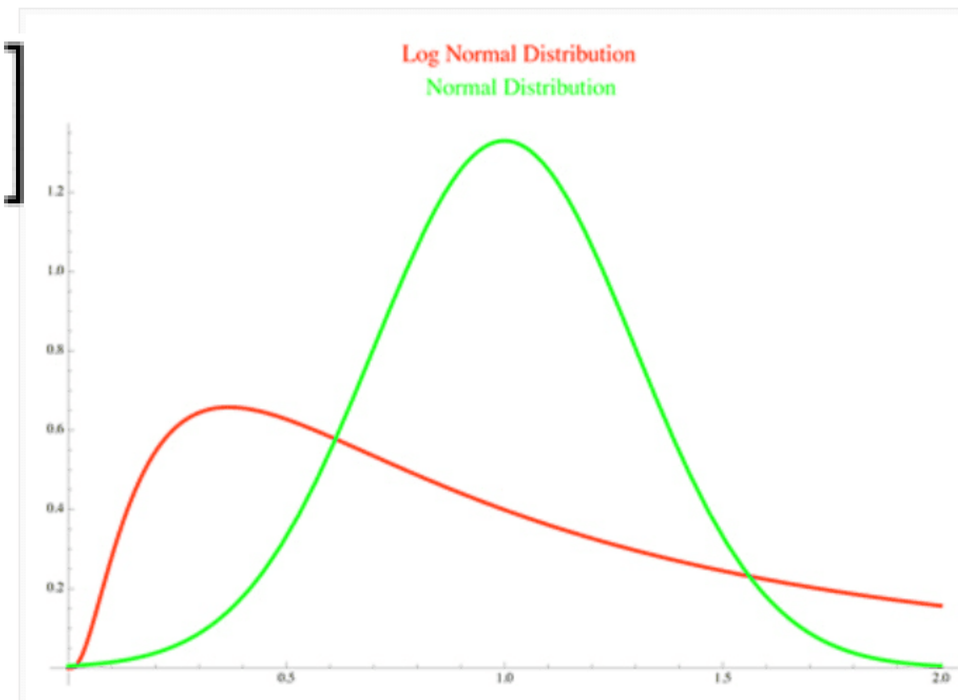
Aerosol Parameters (4)

1. Particle column density (particle density, N_0, particles/cm^3 x Pathlength, cm; N_0L)
2. Composition: wt% sulfuric acid

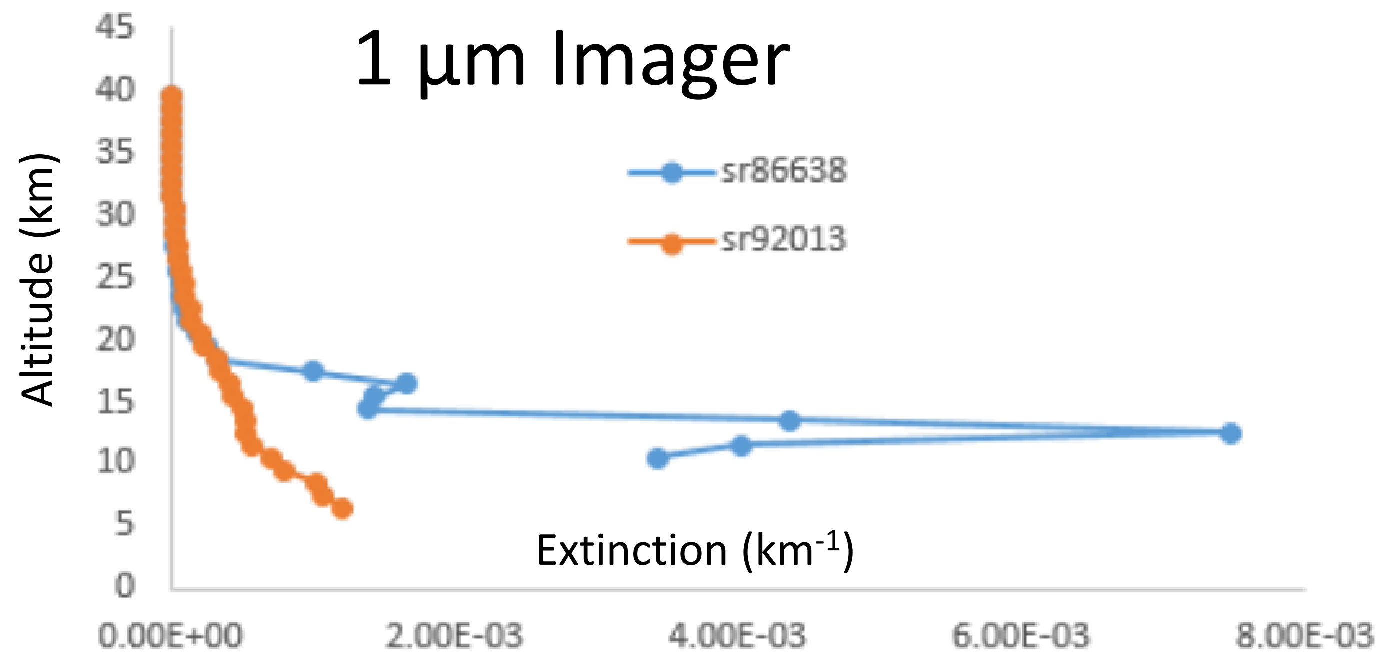
Size distribution: assume lognormal

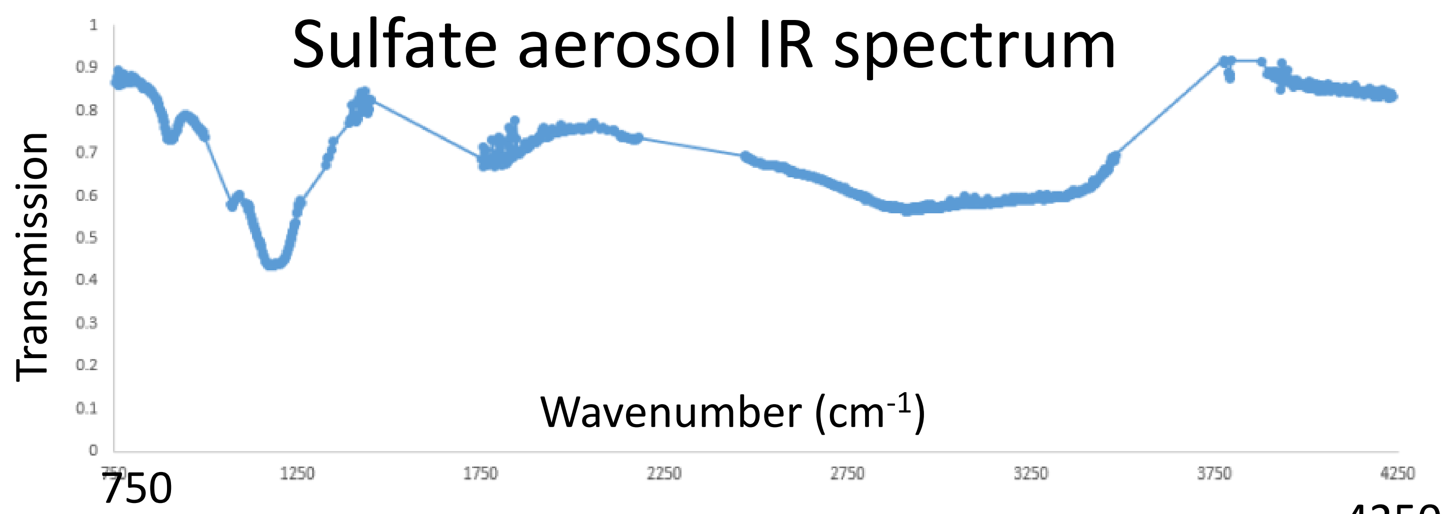
3. r_m: median radius
4. S : spread of distribution (SD in lnr space)

$$n(r) = \frac{N_0}{\sqrt{2\pi}}\frac{1}{\ln(S)}\frac{1}{r}\exp\left[-\frac{(\ln r - \ln r_m)^2}{2\ln^2(S)}\right]$$



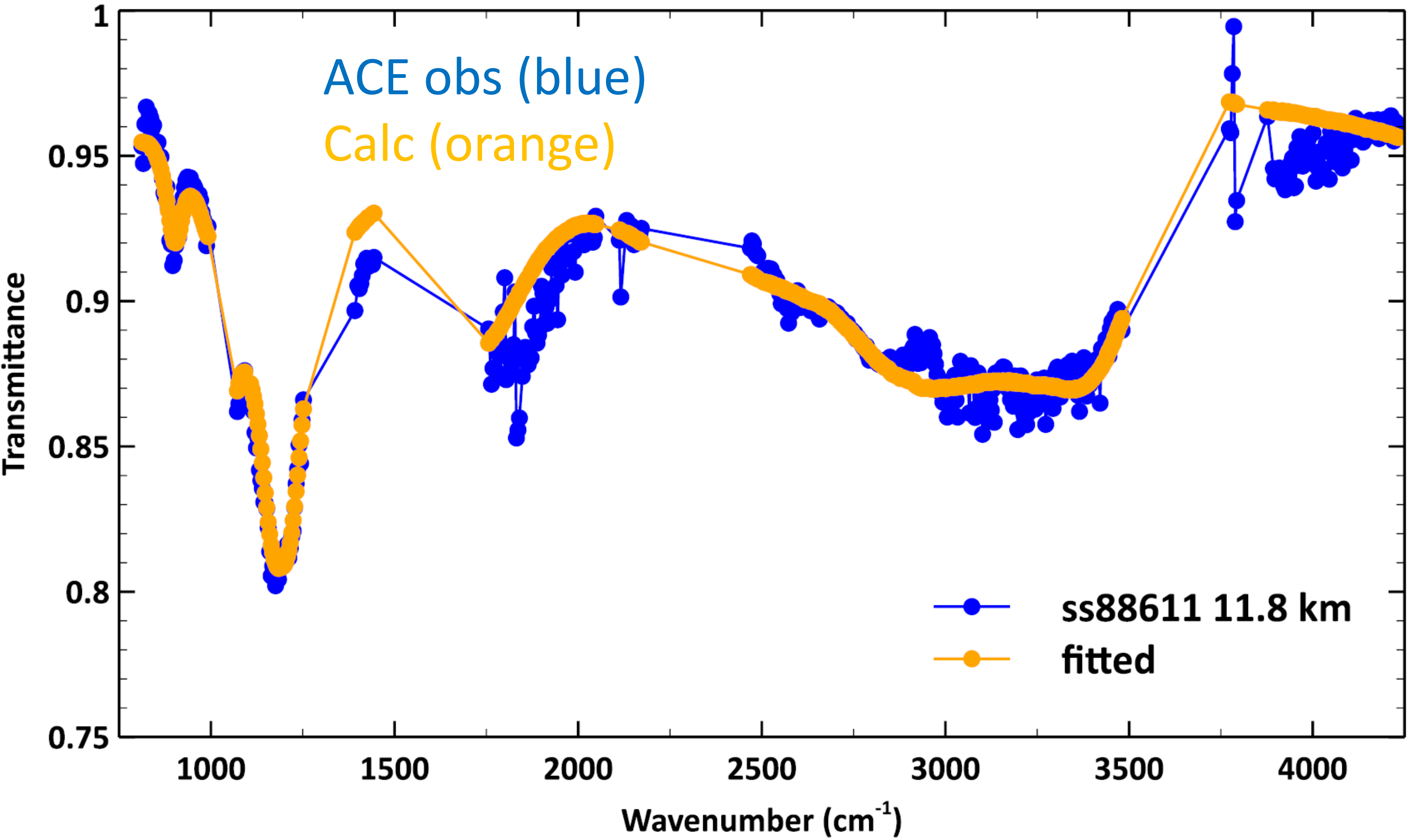
ACE-FTS "Residual" Spectra (Raikoke)





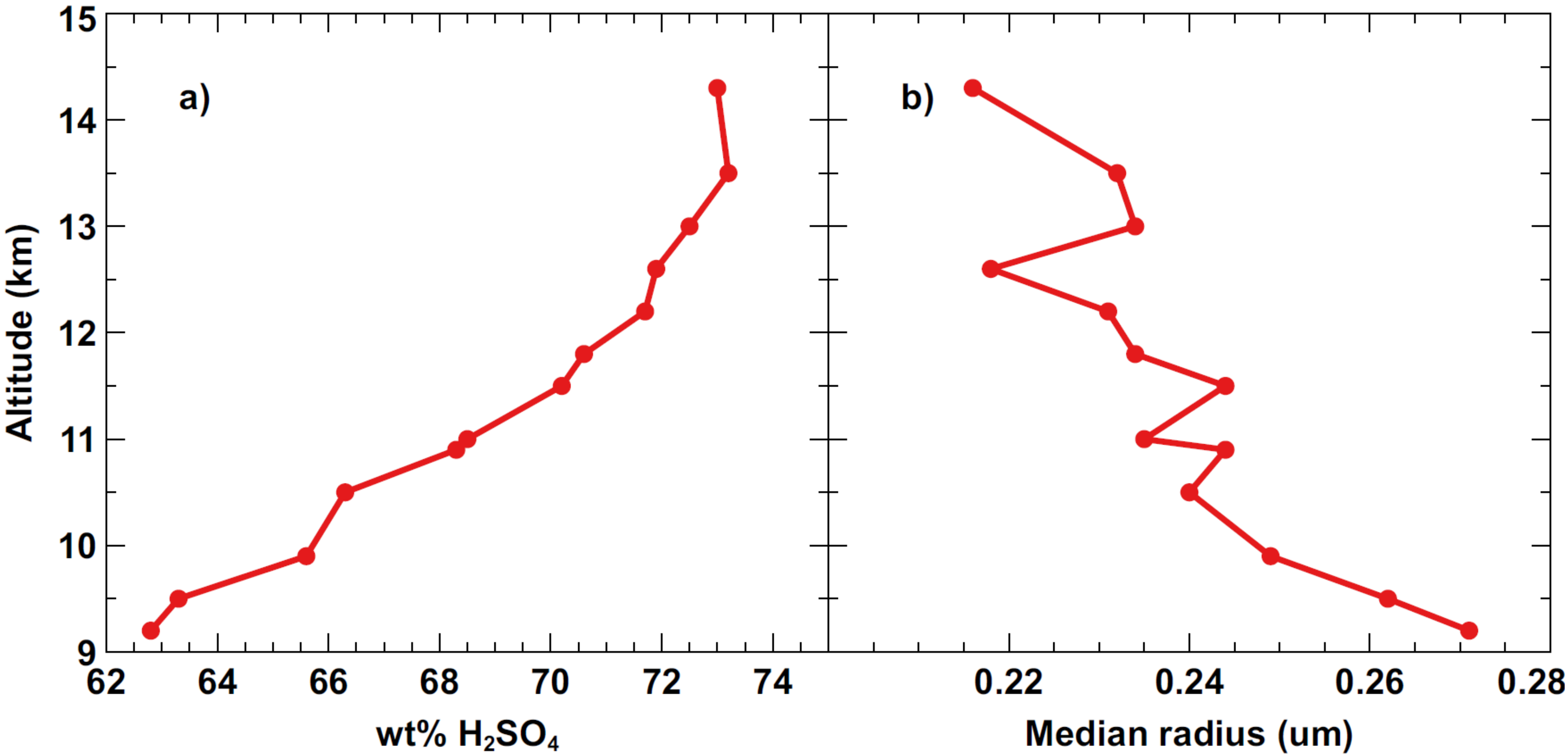
Above: sr86638 11.7 km / sr92013 11.5 km

Sulfate Aerosol Fit



Parameters: 65.3±0.8%(wt), r_m = 0.22±0.05 μm, T = 227 K (fixed), S=1.3 (fixed); 3.4x10^8 particles/cm^2

Change in sulfate particle composition and size with altitude

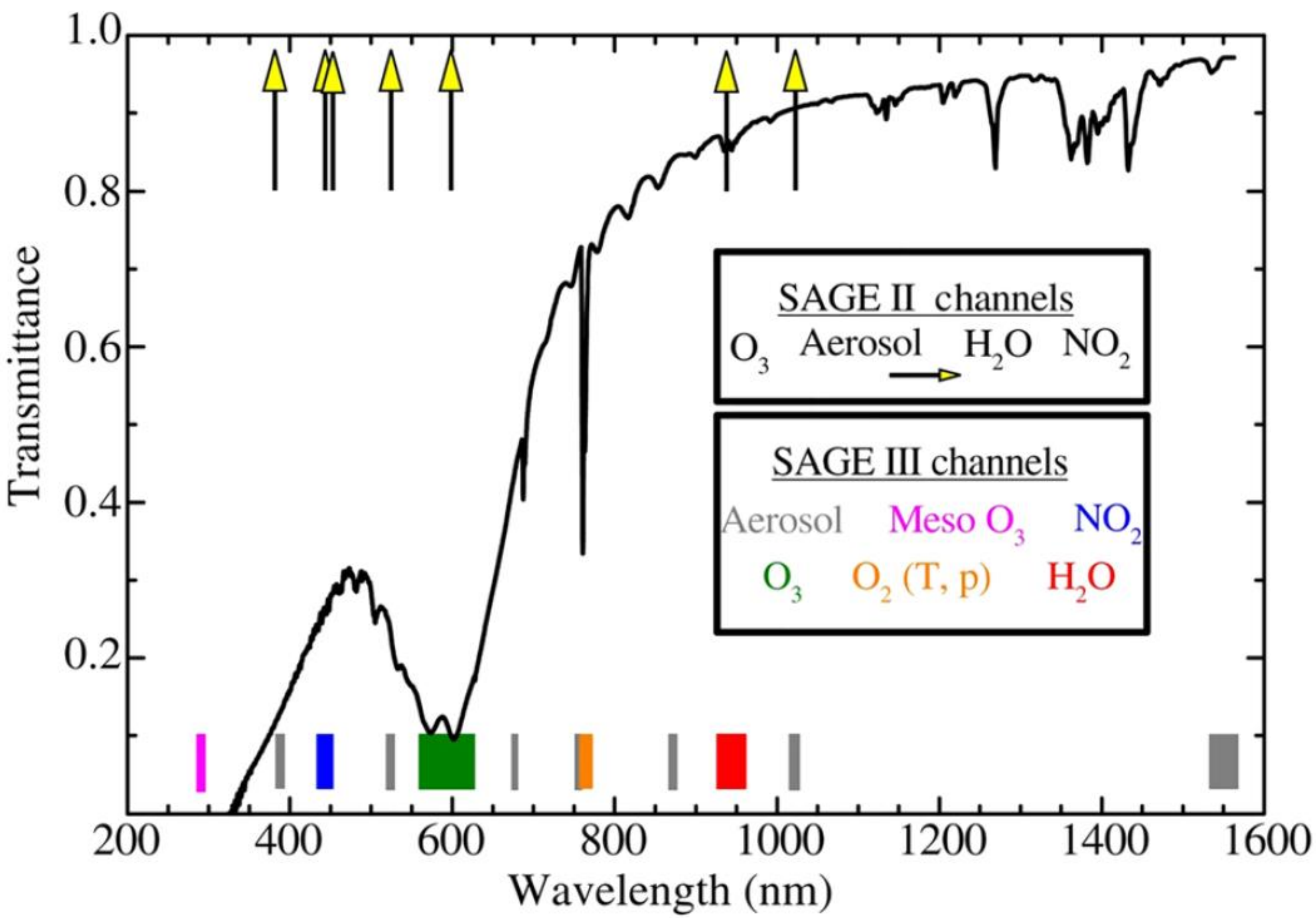


ACE-FTS Sulfate Aerosol Retrievals

- ACE sulfate aerosol spectra are due mainly to absorption with some scattering at higher wavenumbers.
- ACE data alone: composition is well determined; r_m is determined; N_0L, particle column density is determined; distribution width, S, is not determined.
- SAGE-III-ISS aerosol extinctions are mainly scattering and will improve ACE retrievals of r_m and S.

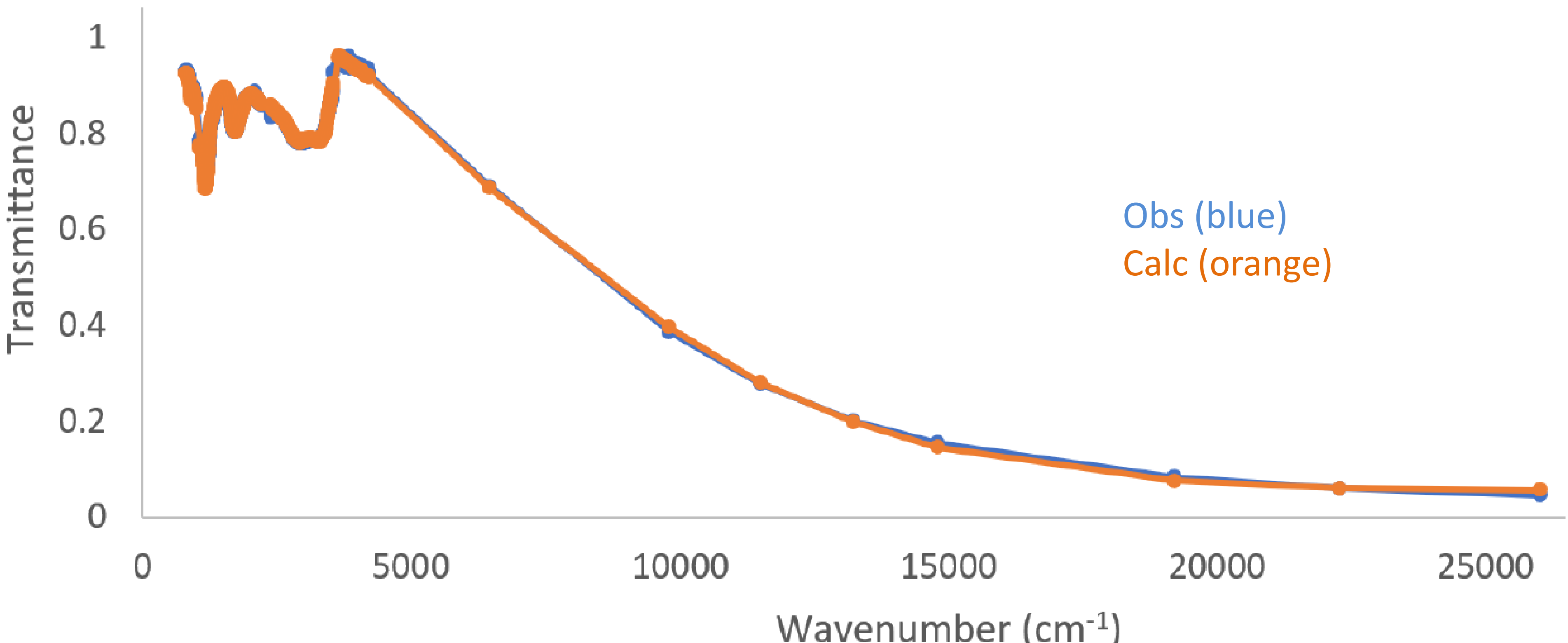
SAGE-III-ISS Aerosol Channels

Chan-nel	λ (nm)
1	384.2
2	484.5
3	520.5
4	601.5
5	676.0
6	755.9
7	869.1
8	1021.2
9	1543.9



ACE-FTS and SAGE-III-ISS

Raikoke plume at 20 km; ACE and SAGE are 2 days apart

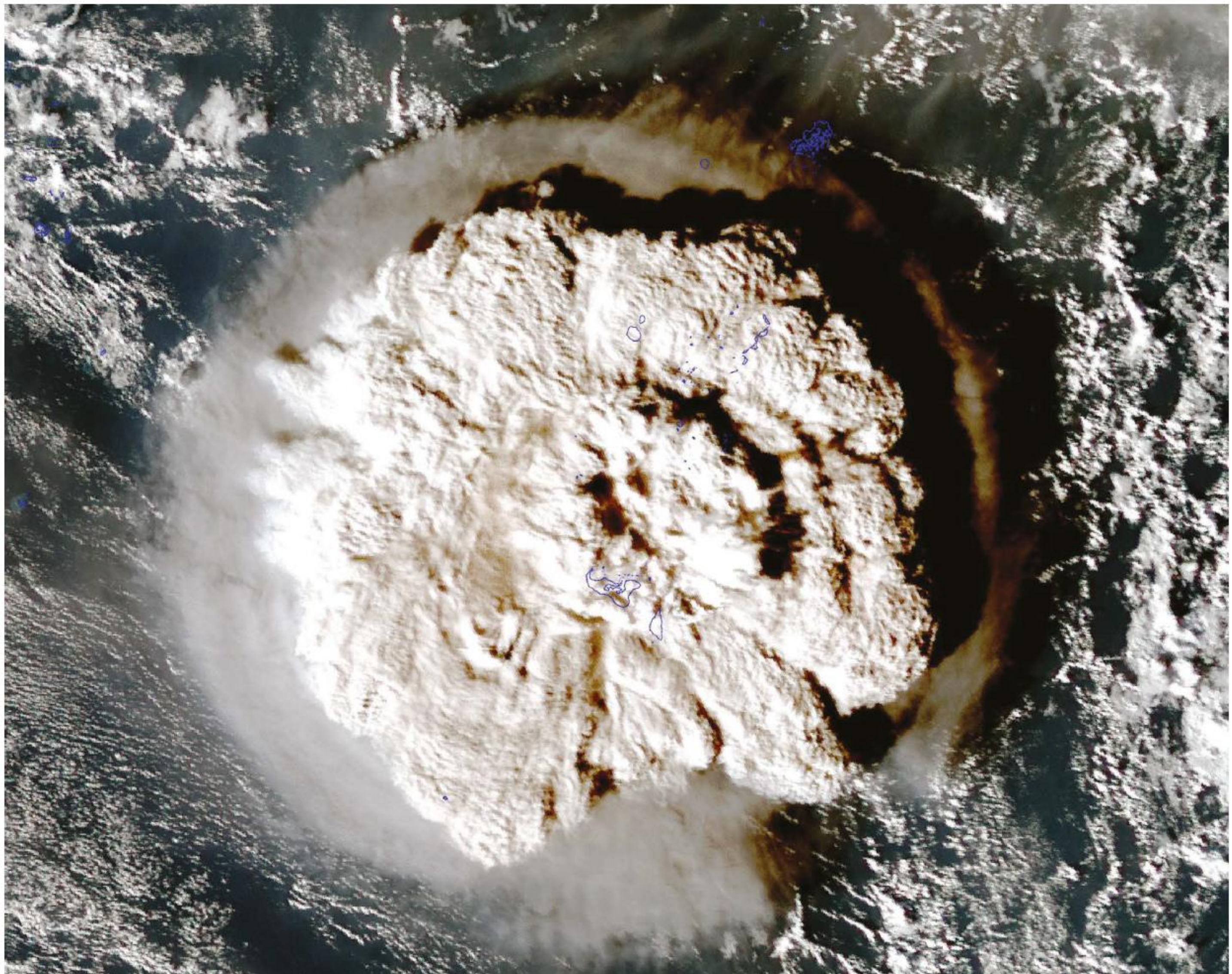


ACE software is used to convert SAGE aerosol extinction values (km^{-1}) into ACE-like limb transmission.

Results: $r_m = 0.241 \pm 0.004$ μm, $H_2SO_4 = 64.9 \pm 0.5$ %, $S = 1.30 \pm 0.01$, $N_0L = 4.8 \pm 0.2$ $\times 10^8$ particles/cm^2

Hunga Tonga–Hunga Ha‘apai Volcano

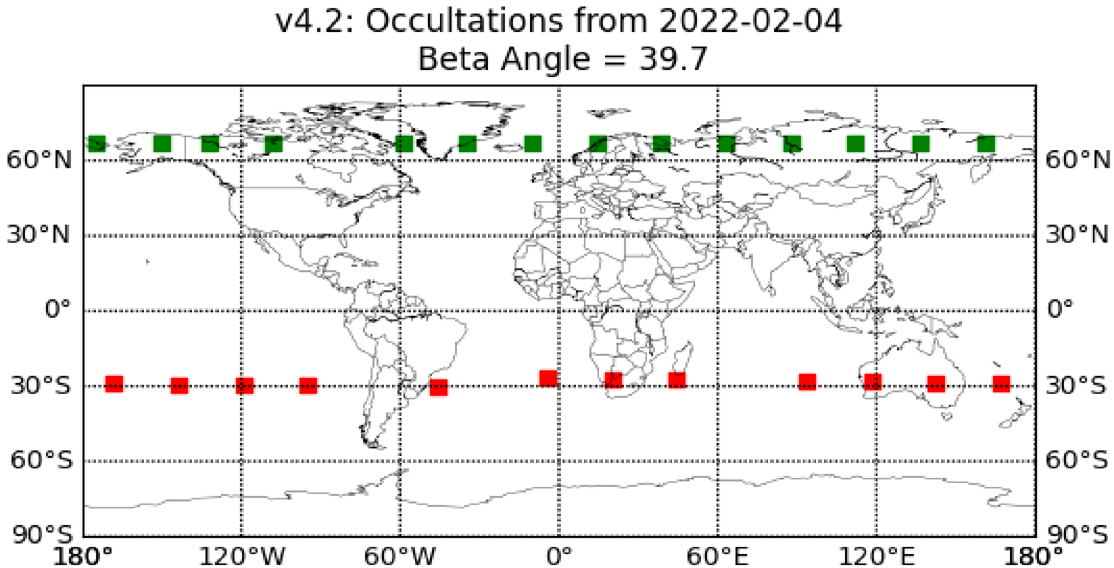
Underwater volcano near Tonga erupted explosively on 15 January 2022: atmospheric pressure and gravity waves, tsunami.



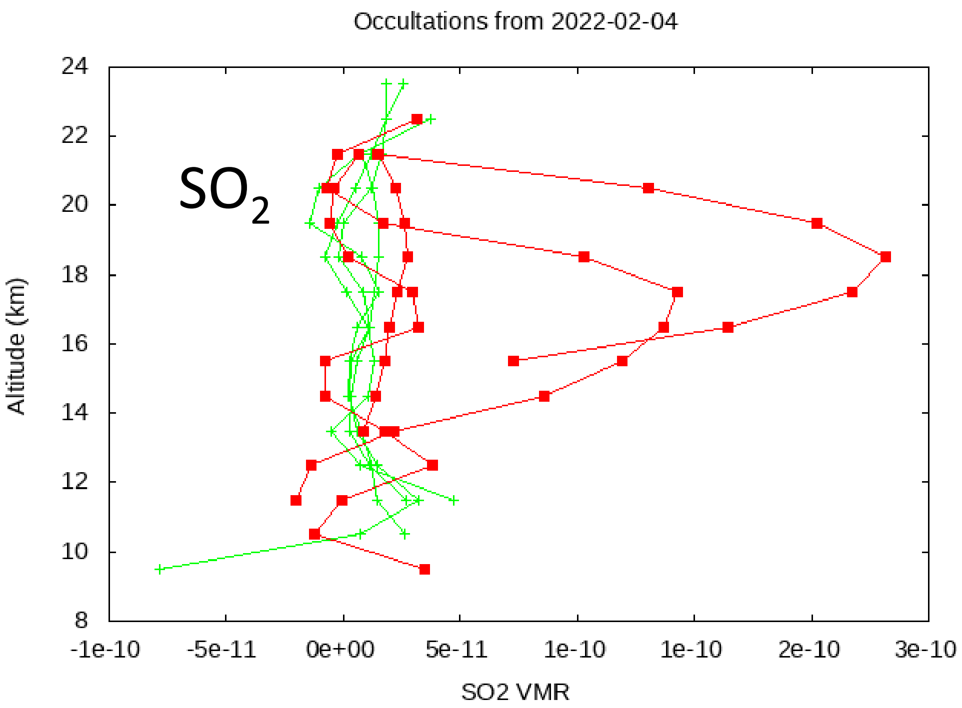
The Japanese satellite Himawari-8 captured the giant ash cloud from the 15 January eruption of Hunga Tonga–Hunga Ha‘apai.

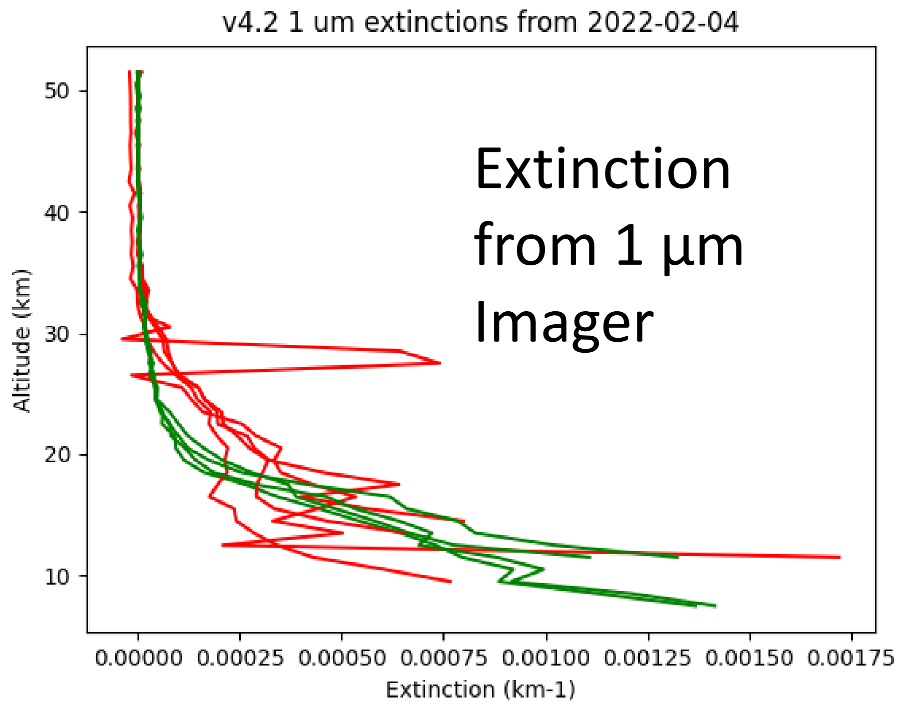
Nature 17 Feb 2022

ACE Observations of Tonga Volcano

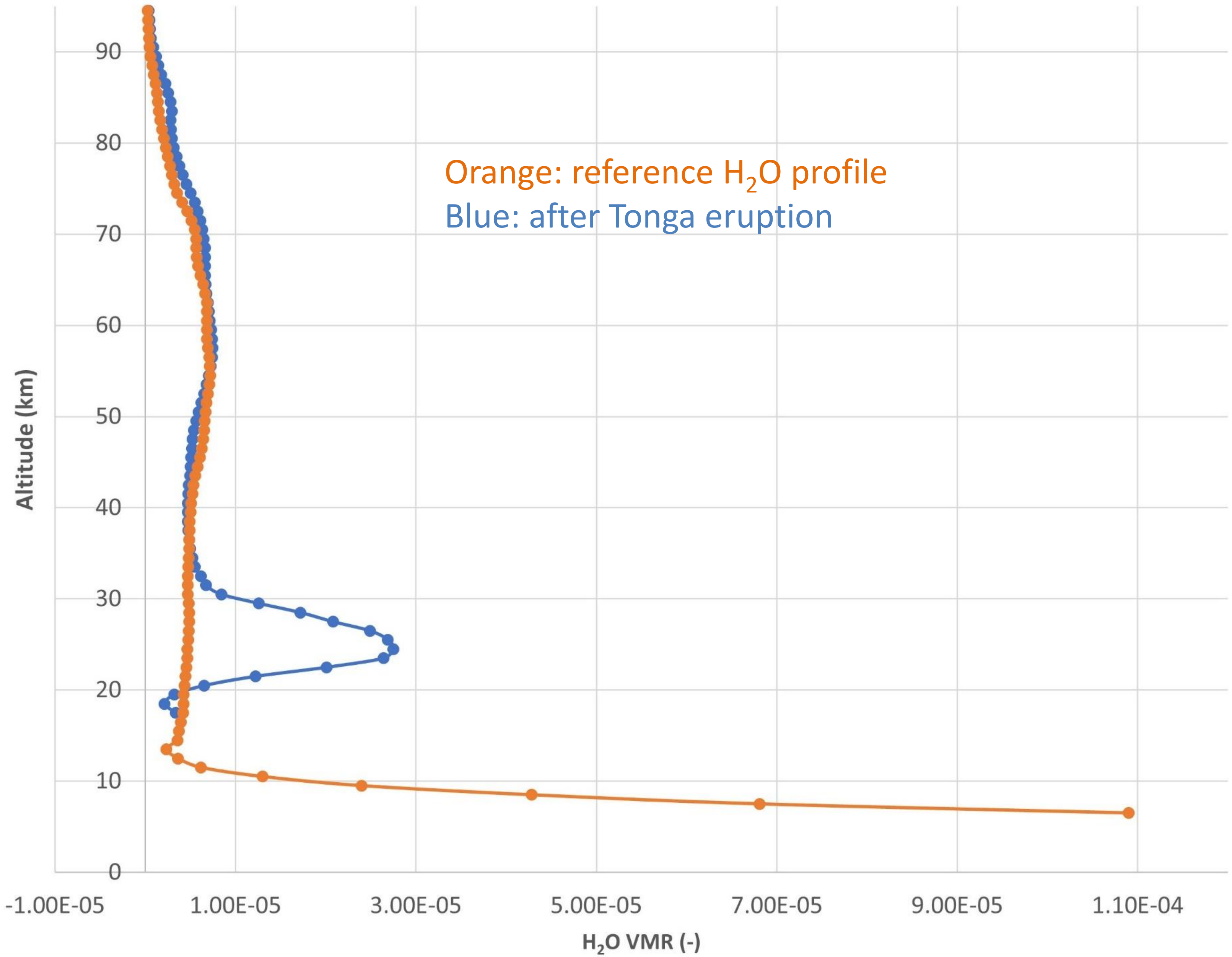


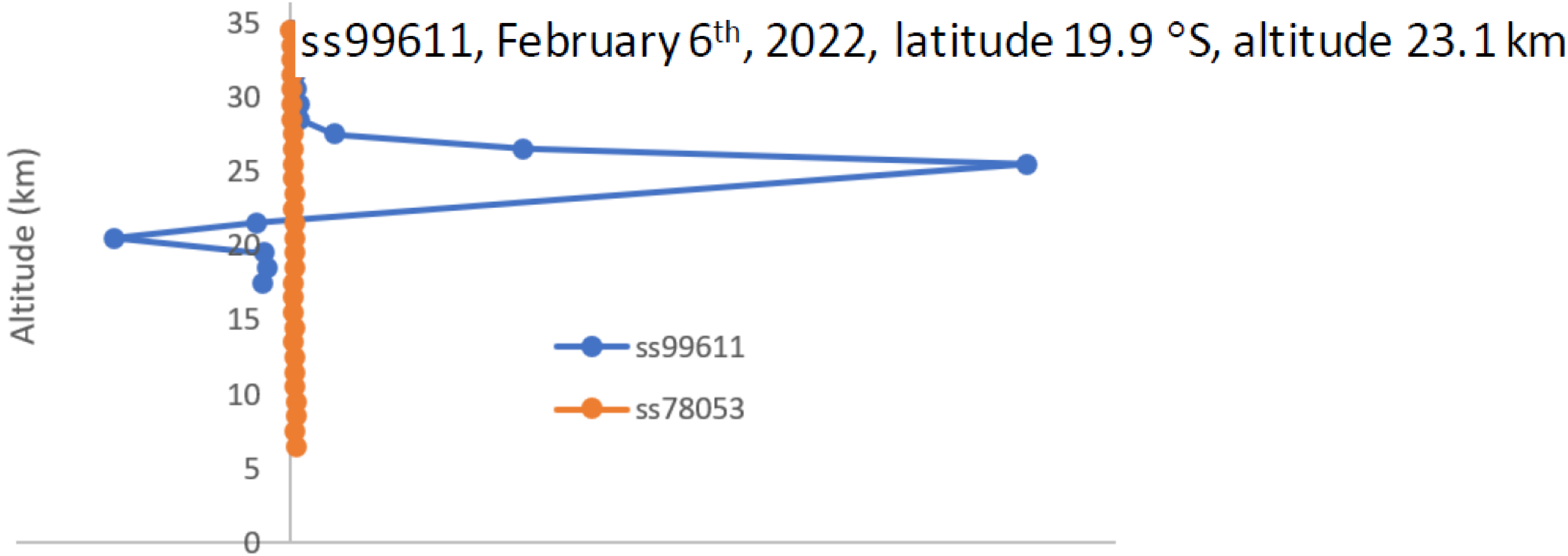
Version 4.1/4.2 UW Near-Real Time Processing for Feb. 4, 2022



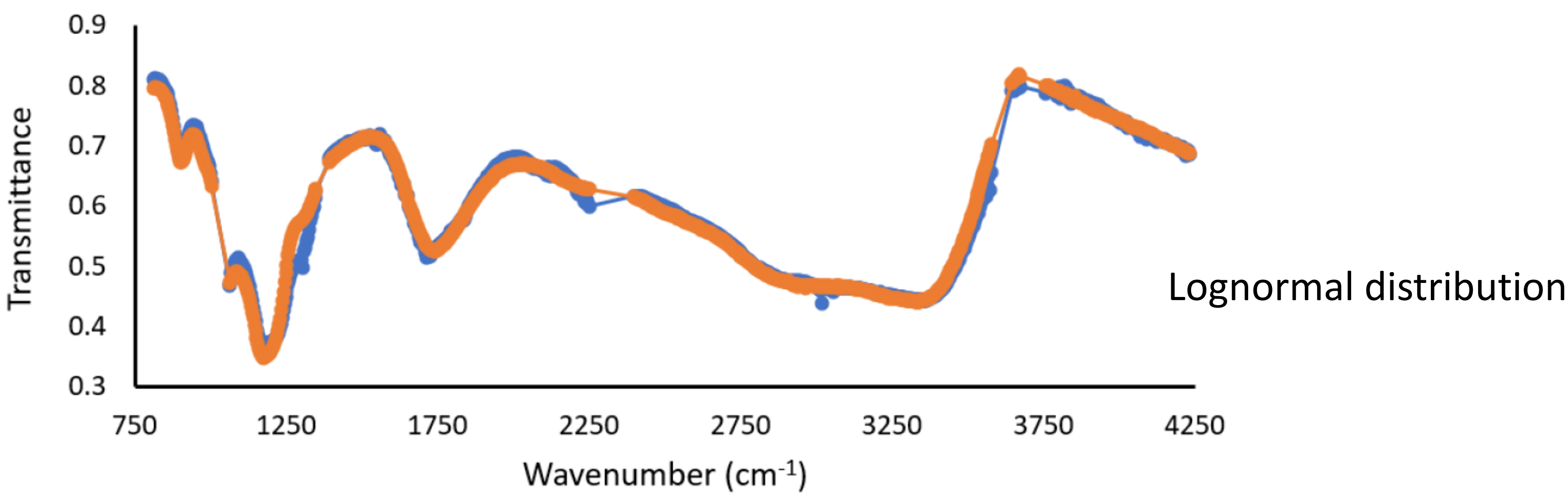


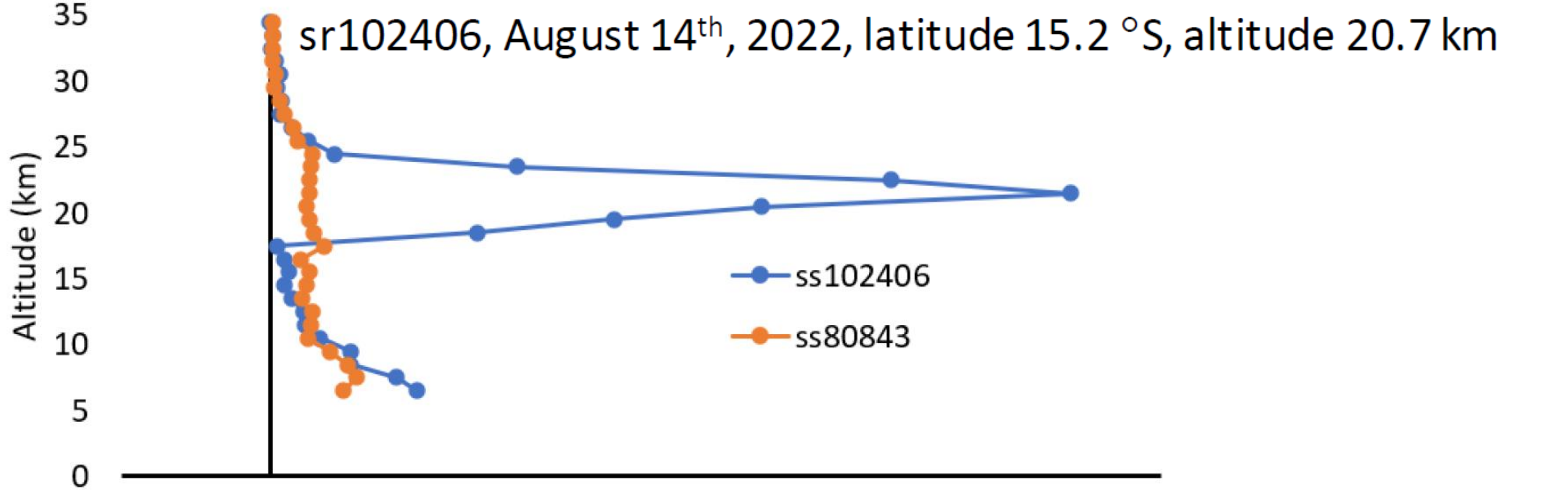
H_2O Profile Feb. 6, 2022



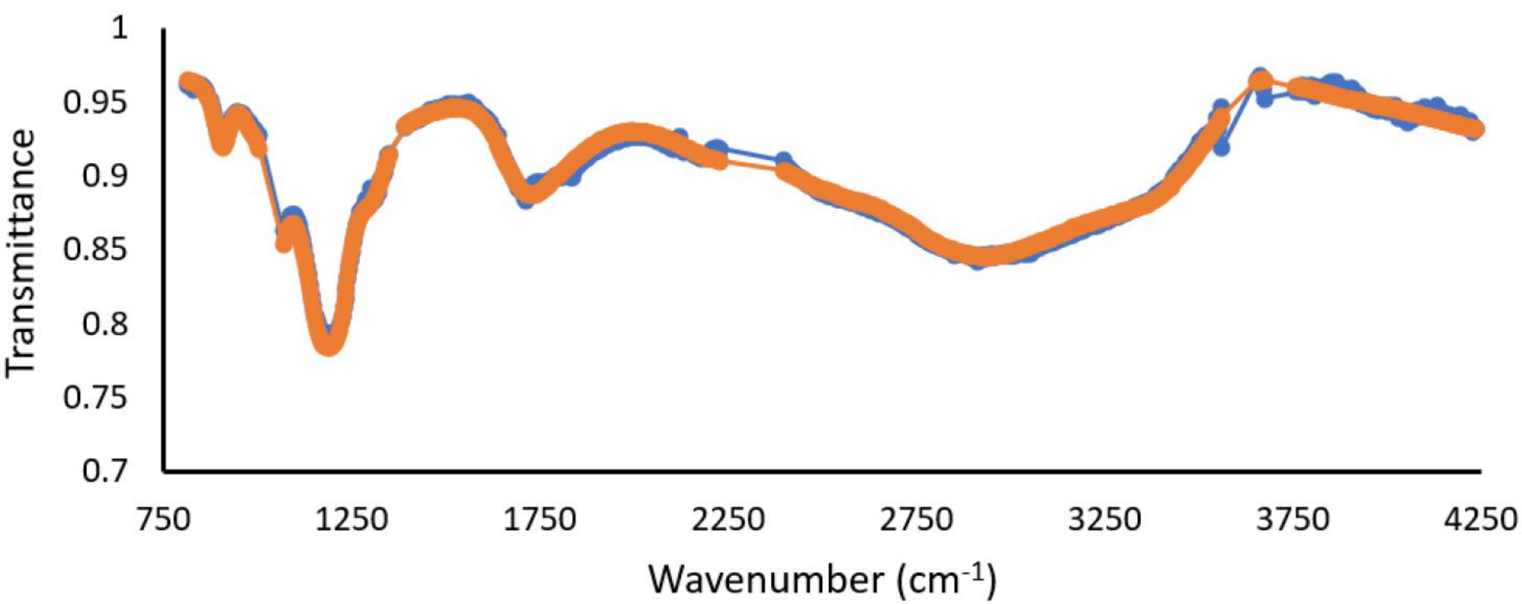


Size = 0.319 ± 0.003 μm, wt% H_2SO_4 = 62.4 ± 0.2, N_oL = (5.3±0.1) x 10^8 particles/cm^2, fixed T = 212.5 K, S = 1.3





Size = 0.315 ± 0.007 µm, wt% H_2SO_4 = 71.2 ± 0.5, N_oL = (1.20±0.08) x 10^7 particles/cm^2, fixed T = 211 K, S = 1.3



Conclusions

- ACE-FTS residual spectra provide a unique dataset for characterization of clouds and aerosols.
- For sulfate aerosols, ACE-FTS data alone provides 3 of 4 parameters (r_m, wt%, N_0L).
- Adding co-incident SAGE-III-ISS data adds S, distribution width, for a complete set of sulfate aerosols parameters, assuming a lognormal distribution.
- ACE aerosol extinctions (version 5.0) derived from 1 micron imager agree with corresponding SAGE-III-ISS values.

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Doug Cameron
Jason Sorensen

It’s All Spectroscopy

Textbook aimed at graduate students and senior undergrads. Particularly useful treatment of the confusing topic of line intensities needed for remote sensing. 4th edition (April 2020) includes atmospheric and astronomical spectroscopy.