The Role of H_2S in Archean Organic Haze Chemistry

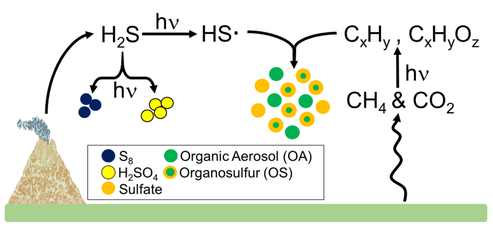
Nathan W. Reed, Margaret A. Tolbert, Eleanor C. Browne

INTRODUCTION

- **Methane-produced organic haze, CO_2, and volcanic/biological H_2S were likely present in the Archean atmosphere.**
- **Current view of Archean atmosphere separates their respective chemistries.**
- **Organic haze production is currently thought to decrease with high CO_2 to CH_4 concentration ratios (greater than 1:1).**

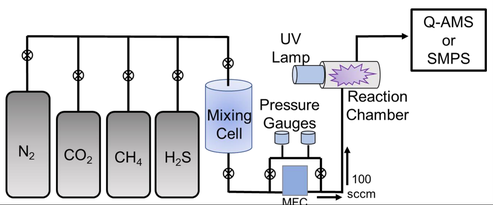
CURIOUSITIES EXPLORED

- **What is the interplay between H_2S and haze formation chemistry? How does this change as a function of CO2?**
- **Could this interplay impact climate, habitability, or atmospheric chemistry of the Archean Earth?**

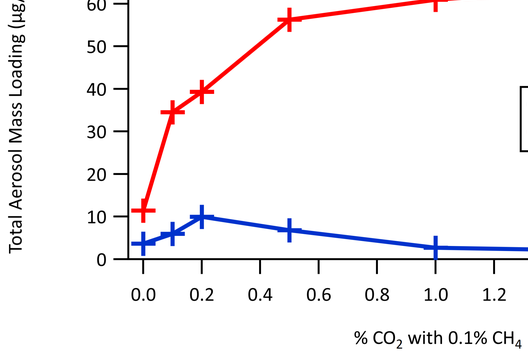


METHODS

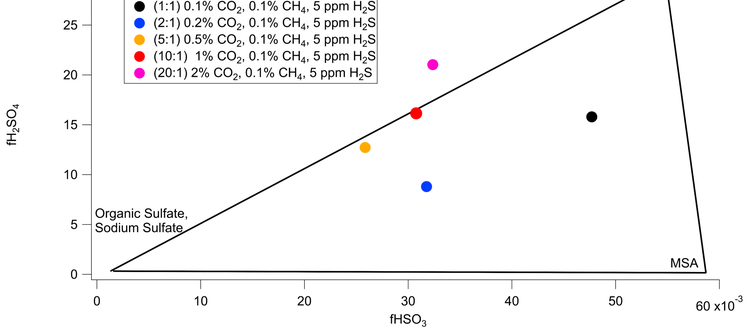
- **Generate haze particles with trace amounts of H_2S via a flow system and UV reaction cell.**
  - **Analyze with quadrupole aerosol mass spectrometry (Q-AMS) or Scanning Mobility Particle Sizer (SMPS).**
- **Measure the particle composition, size, and number in real time.**



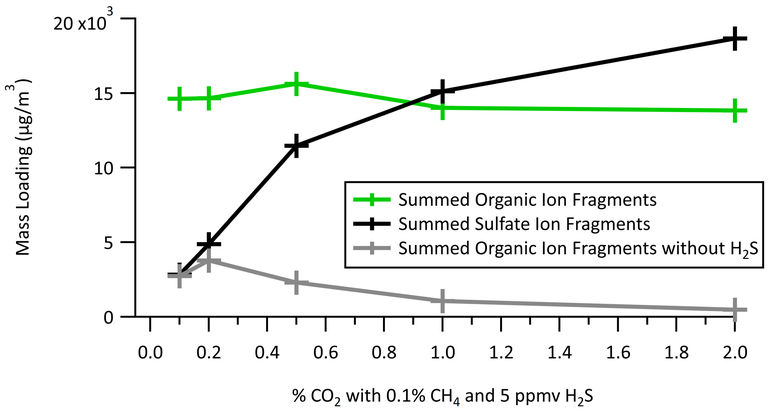
Laboratory experiments showed trace H_2S ***increased*** organic aerosol production in high-CO_2 organic haze chemistry. Signs of the formation of organosulfur and sulfate aerosol were also observed.



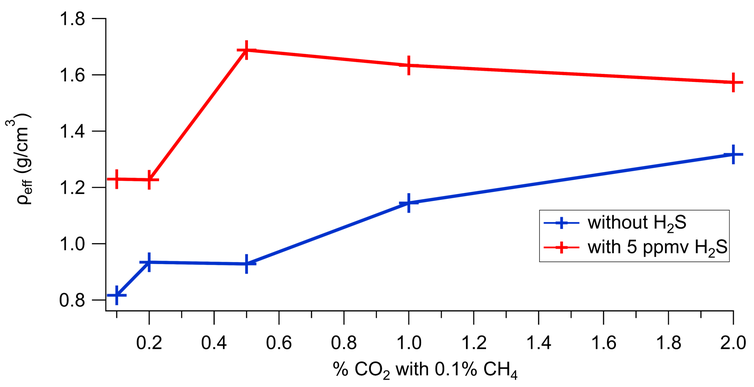
- **With 5 ppmv H_2S, the total aerosol mass loading increases with increasing initial CO_2 concentration**



- **Standard sulfate fragmentation patterns could imply a mixture of sulfate and organosulfate aerosol**



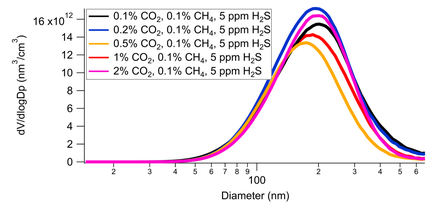
- **Including 5 ppmv H_2S produces sulfate fragments, but the amount of organic fragments remain relatively constant**

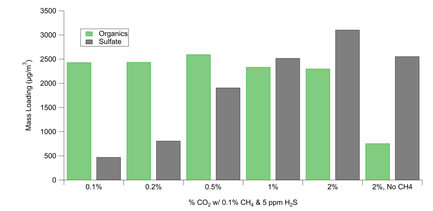


- **Particle density is higher with H_2S and initially increases with CO_2, but slightly decreases after 0.5% CO_2**

CONCLUSIONS & SIGNIFICANCE

- **Trace amounts of H_2S (5 ppmv) in Archean-like gas mixtures produced organic and sulfate aerosol, *even at higher CO_2: CH_4 concertation ratios.***
- **There was no S_8 found at each CO_2 concentration, and only *low H_2SO_4* at higher CO_2 concentrations.**
- **Evidence leaned toward *organosulfur* and *organosulfate* aerosol.**
- **These results *defer from the current thought* of Archean atmospheric sulfur reservoirs.**
- **This may be significant for interpreting the sulfur isotopic records, understanding the Archean atmosphere/climate and the early evolution of the atmosphere.**





1. University of Colorado Boulder & Cooperative Institute for Research in Environmental Science (CIRES), Boulder, CO 80309

Trainer, *PNAS*, **2006**, 103, 18035-18042; DeWitt, *Astrobiology*, **2010**, 10, 8, 773-781; Kasting, *Science*, **2001**, 293, 819-820; Farquhar, *Earth and Planet. Sci. Lett.*, **2003**, 213, 1-13; Domagal-Goldman, *Earth and Planet. Sci. Lett.* **2008**, 269, 29-40; Arney, *Astrobiology*, **2018**, 10.1089/ast.2017.1666; Hu, *Astrophysical Journal*, **2013**, 769, 6; Kump, *Nature*, **2007**, 448, 1033-1036; Bates, *Atmos. Chem. Phys.*, **2006**, 6, 1657–1732; Horst, *Astrobiology*, **2012**, 12, 9, 809-817; Horst, *The Astrophysical Journal Letters*, **2013**, 710:L10, 1-6S; Reed, *ACS Earth and Space Chem*, **2020**, 4, 897-904

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