

Supporting information for:

Rapid, concurrent formation of organic S and iron sulfides during experimental sulfurization of sinking marine particles

M. R. Raven^{1*}, R. G. Keil², S. M. Webb³

¹ Department of Earth Science, University of California, Santa Barbara, Santa Barbara CA 93117, USA

² School of Oceanography, University of Washington, Seattle WA 98195, USA

³ Stanford Synchrotron Radiation Lightsource, Stanford University, Menlo Park CA 94025, USA

Contents of this file:

- Figure S1** Sample processing workflow diagram
Figure S2 XAS standards used in fits
Figure S3 Molar N:C ratio of particle materials
Figure S4 XAS spectra for ETNP particles

Tables (see .xlsx file)

- S1** Sample IDs and collection details
S2 Carbon isotope and elemental compositions of particle materials
S3 Sulfur isotope data
S4 XAS fits, macro (500 μm^2) spots for bulk speciation
S5 XAS fits, micro (1 μm^2) spots for map speciation

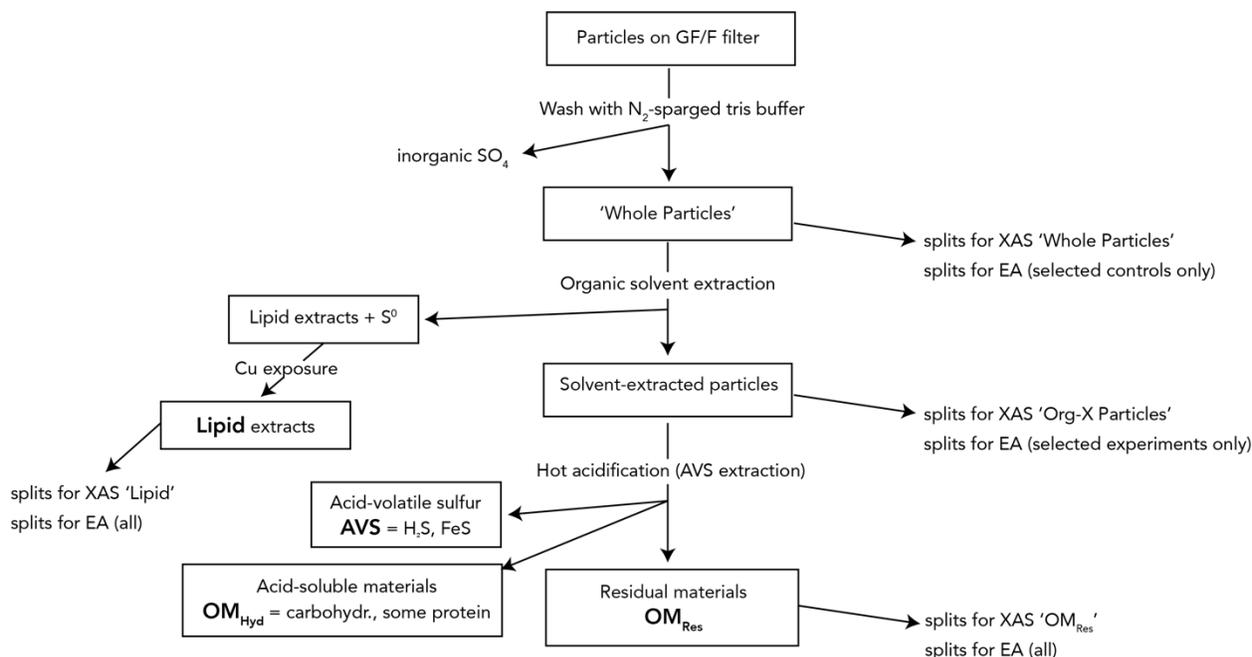


Fig. S1 **Sample processing workflow diagram.** Particles were sequentially extracted with buffer, organic solvent, and strong acid; splits were collected after each step for X-ray absorption spectroscopy (XAS) and elemental analysis (EA). Pool IDs correspond to those in Fig. 2 and throughout the main text: OM_{Res} = residual, acid-resistant organics; OM_{Hyd} = hydrolyzable and/or acid-soluble organics; AVS = acid-volatile sulfides.

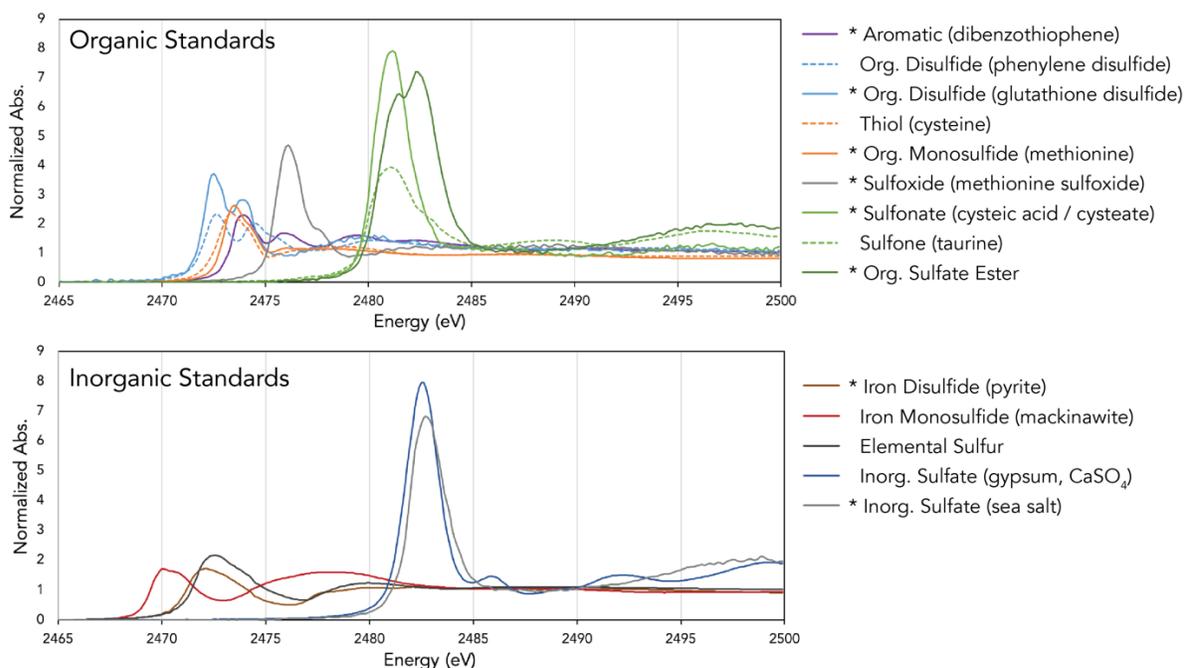


Fig. S2 XAS standard spectra used in fits. Standards are curated from a collection of in-house results (processed identically to samples, denoted by ‘*’) and the European Synchrotron Radiation Facility database (<https://www.esrf.fr/home/UsersAndScience/Experiments/XNP/ID21/php.html>), unstarred. All standard spectra are calibrated to a consistent monochromator energy (eV) relative to our thiosulfate pre-edge peak at 2472.02. Linear combination fits were calculated using multiple examples of some categories of organic sulfur structures to provide coverage of the real variability within natural materials (dashed and solid lines of the same color indicate organic monosulfides and thiols; sulfones and sulfonates; two examples of organic disulfides). These spectra were summed as single bins during all subsequent data analysis.

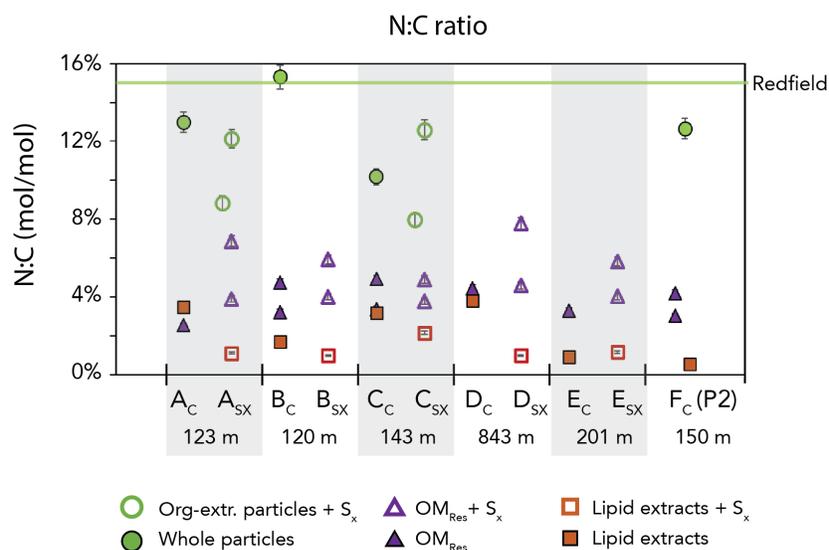


Fig. S3 Molar N:C ratio of particle materials, before and after S_x exposure. Filled symbols represent controls (e.g., ‘A_C’), and hollow symbols represent experiments (e.g., ‘A_{SX}’), as detailed in Table S1. Whole and organic-solvent-extracted particles contain both OM_{Hyd} and OM_{Res}. Samples with multiple symbols represent discrete filter splits rather than duplicates of homogenized samples. Error bars indicate the long-term reproducibility of standards (2σ).

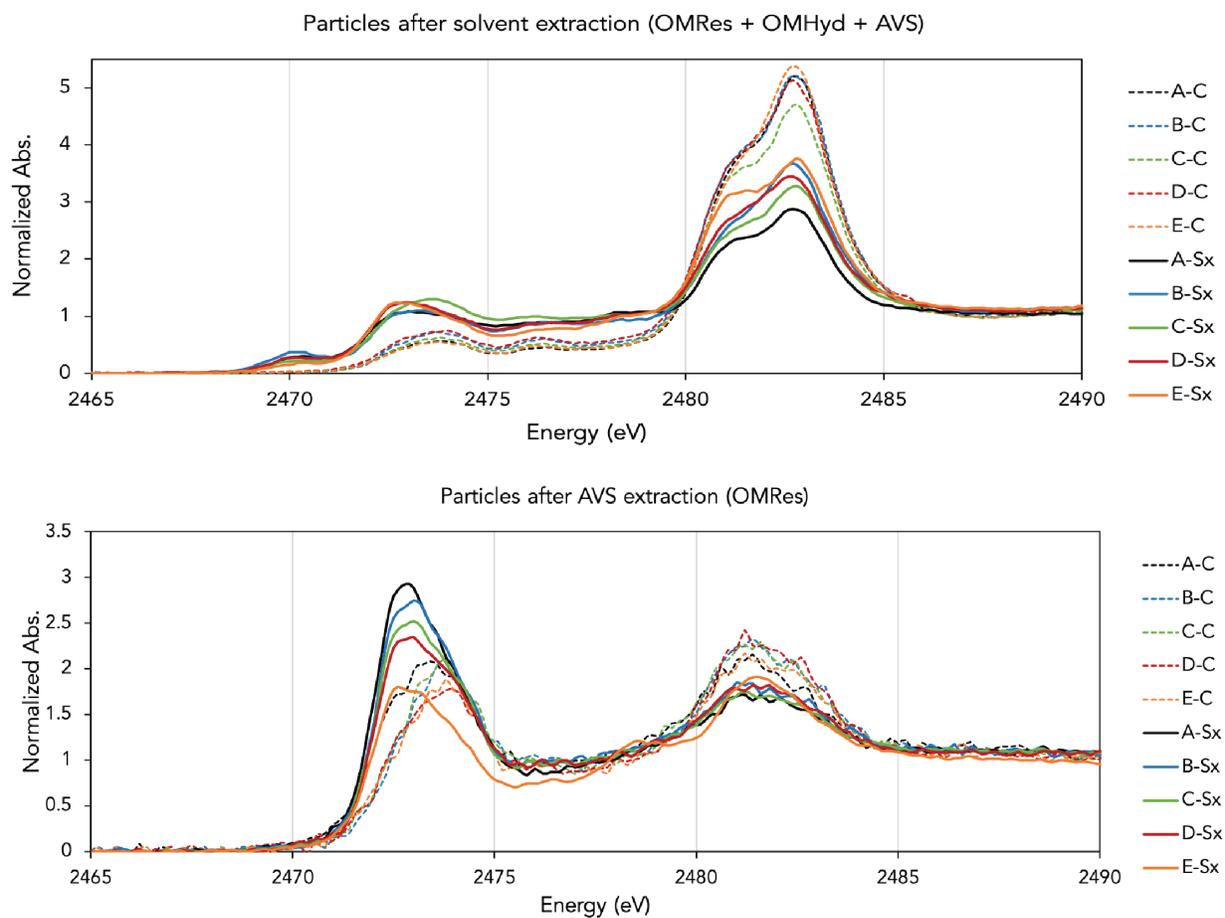


Fig. S4 XAS spectra for ETNP particles. All spectra are normalized to a post-edge baseline of 1.0 using pre- and post-edge normalization regions of -20 to -7 and +35 to +60 eV, respectively, relative to an E0 of 2473.0 eV. Dashed lines show controls, and solid lines show materials after polysulfide exposure. Top: Particle materials after solvent extraction but prior to acidification, including acid-resistant (OM_{Res}), acid hydrolyzable (OM_{Hyd}), and acid-volatile (AVS) sulfur species. These spectra were used to calculate newly formed materials (c.f. Fig. 6). Bottom: Particle materials after solvent extraction and subsequent hot acidification (AVS extraction), defined as OM_{Res} .