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Expanding the View of Wildfire at the Paleocene-Eocene Boundary: A Novel Molecular Marker Approach

SASHA WAGNER[1], MORGAN SCHALLER[1], RILEY BARTON[1], SEBASTIAN BARKETT[1], CELESTE CORCORAN[1] AND MEGAN FUNG[2]

[1]Rensselaer Polytechnic Institute

[2]California Lutheran University

Presenting Author: wagnes3@rpi.edu

Wildfires produce charcoal and soot that varies in size, mobility, composition, and reactivity, complicating the interpretation of pyrogenic signals recorded in ancient sediments. Depending upon the method used, significant fractions of sedimentary pyrogenic carbon are "invisible" to detection. For example, the quantification of particulate charcoal is visually compelling, but provides a somewhat narrow view of paleo-wildfire events and their global impact because fine residues, which comprise a significant portion of the pyrogenic material left behind, are excluded. The soot-like carbon reservoir is accessed through analysis of benzenepolycarboxylic acid (BPCA) molecular markers, which quantify a condensed aromatic form of pyrogenic carbon in sediments that is not represented by particulate charcoal alone [1]. Here, we show BPCAs to be a strong indicator of paleo-wildfire at the Paleocene-Eocene (P-E) boundary (~56 Ma), which is marked by the onset of a global carbon isotope excursion (CIE) and rapid ~5°C warming and is thus cited as an analog for anthropogenic climate change. The abrupt Paleocene-Eocene Thermal Maximum (PETM) warming is linked to a massive injection of isotopically light carbon into Earth's exchangeable carbon reservoirs that is recorded as a CIE in contemporaneous marine sediments. Particulate charcoal was found in a prominent peak above background levels along the paleo-continental shelf [2]. The sedimentary distribution of BPCAs overlaps with, but peaks after, the deposition of particulate charcoal. BPCA-specific carbon isotope ratios ($\delta^{13}C$) mirror particulate charcoal $\delta^{13}C$ values and indicate burning and mobilization of pre-CIE vegetation. These findings show BPCAs to be a powerful tracer for paleo-wildfire at a critical juncture in Earth's climate history and expand the analytical window for detecting pyrogenic residues in the geologic record. The transfer of particulate charcoal via rivers to the coast is largely constrained by rainfall and hydrologic connectivity, whereas soot-like residues are environmentally ubiquitous, even during infrequent fire periods, owing to their refractory nature and long terrestrial and atmospheric residence times. The apparent difference in maxima between the two wildfire proxies may thus reflect different depositional modes for BPCAs and particulate charcoal.

[1] Wagner et al., (2017) *Limnol. Oceanogr.: Methods* doi:10.1002/lom3.10219

[2] Fung et al., (2019) *Geochem. Persp. Let.* doi:10.7185/geochemlet.1906