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Redox Transformations in Wildfire Ashes addressed by Synchrotron-based X-ray Fluorescence Microprobe Techniques

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Micro X-ray fluorescence (u-XRF) imaging and micro X-ray absorption fine structure spectroscopic (u-XAFS) analysis of wildland-urban interface (WUI) fire ashes collected from the LNU Lightning Complex Fire perimeter in Northern California, USA have been performed to evaluate changes in metal(loid) speciation (*e.g.,* oxidation states of Cr, Mn, As, and Ti) speciation as a result of combustion processes. These experiments were conducted at beamlines 4-BM at the National Synchrotron Light Source II (Brookhaven National Laboratory) and 13-ID-E at the Advanced Photon Source (Argonne National Laboratory). The u-XRF elemental mapping allowed identification of high and low concentration spots of the different metal(loid) species and their oxidation states. Our findings demonstrate that a mixture of both oxidized and reduced species is present in the WUI fire ashes. Detected oxidized species include Ti(IV), Cr(VI), As(III), and Mn(IV). In contrast to reduced species as Ti(III), Cr(III), As(III), and Mn(II). The spatially resolved X-ray microprobe analyses provide a comprehensive picture of the end-member species stabilized and their interrelationship that was not apparent using bulk XAFS spectroscopy. The u-XAFS analysis allows us to establish reference spectra that can then be used for linear combination fitting analysis of the bulk XAFS measurements. Overall, these results demonstrate that complex transformations of metal(loid)s in WUI fire ashes. In particular the reduction of TiO_2 to the substoichiometric Magneli phases (Ti_nO_{2n-1}), As(V) to As(III), Mn(V) to Mn(II) and the oxidation of Cr(III) to Cr(VI). These metal(loid) transformations help explain the higher toxicity of WUI fires compared to particulate matter from other sources such as ambient urban particulate matter.