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Enrichment Factors of Perfluoroalkyl Anionic Surfactants at the Air/water Interface

M. R. Hoffmann, A. J. Colussi, and J. Cheng

California Institute of Technology, Environmental Science & Engineering, Pasadena, CA, United States (mrh@caltech.edu)

The refractory, water-bound perfluoro-$n-$alkyl carboxylate (n-PFC-) and sulfonate (n- PFS-) surfactants reach remote locations hydrologically unconnected to their sources via hitherto unknown processes. Here we quantify the enrichment of these anions in microdroplets produced by aerosolization of 1 μM aqueous surfactant solutions at pH 7 using sonic spray ionization mass spectrometry. Relative enrichment factors $f$steeply increase from n= 1 to 3, level off at $f(n$-PFS-) $\sim$ 2.3 $f(n$-PFC-) $\sim$ 190 f(Br-) ($n \geq 8$), and display even-odd effects. The preferential enrichment of n-PFS- over n-PFC- reflects a larger headgroup. The hyperbolic, rather than the predicted linear ln $f$vs. $n$dependence suggests the onset of conformational restrictions to interfacial enrichment above $n \sim 3$. Marine aerosols produced from contaminated ocean surface waters are thus expected to be highly enriched in PFC-s/PFS-s. The monotonic accumulation of 8-PFS- and PFC-s in East and, less pronouncedly, in West Greenland biomarkers is therefore consistent with the deposition of marine aerosol drawn from contaminated North American Current waters and transported westward by predominant easterlies. The analogous trend for PFC-s in Canadian Arctic biota vis-a-vis the rapid decline of 8-PFS- after 2000 suggest that the fresher Labrador Sea does not sustain a similar process over this region.