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Uranium-natural organic matters complexation constants determination by spectrofluorescence quenching

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Uranium (U) is an actinide, both radio- and chemio-toxic, present in trace amounts in the environment. Its bio-availability, which is directly linked to its chemical speciation, plays a key role in assessing its potential risks to people and environment. In aqueous systems, dissolved organic matter (DOM) significantly influences the U speciation. However, despite numerous studies on U behavior, the role of DOM in this process remains insufficiently characterized. Currently, geochemical models rely on limited thermodynamic databases, with the only available U-DOM complexation constants being those provided by Milne et al. (2001)[1]. These constants are derived from poor data sets, which limits the accuracy of speciation predictions given the natural variability of DOM across aquatic environments. DOM in freshwater systems is generally composed of proteins, humic, and fulvic acids, whose relative proportions differ for each DOM sources, leading to variations in DOM complexation behavior.

This study aimed at characterizing U-DOM interactions by quantifying the reactive sites involved in U complexation and determining their associated complexation constants. Eight DOMs (5 humic acids, 1 fulvic acid, and 2 aquatic natural organic reference matters) were examined under environmental pH conditions (4, 6, 7, and 8). Identification of proteic, humic and fulvic-like poles in each DOM was done using 3D spectrofluorescence. U-DOM complexation constants were subsequently determined through fluorescence quenching experiments following the gradual addition of U.

Results indicated that DOMs contain two main reactive poles (humic- and fulvic-like) in varying proportions (Figure 1). The apparent complexation constants were obtained by fitting a model to the fluorescence quenching curves of each DOM reactive pole, vary depending on the type of DOM and pH (Figure 2). This work will present results using a more comprehensive model developed to incorporate competition between the poles and interactions with H^+, aiming to refine the determination of true complexation constants.

[1] Milne et al, 2001. Environ. Sci. Technol. 35, 2049–2059.

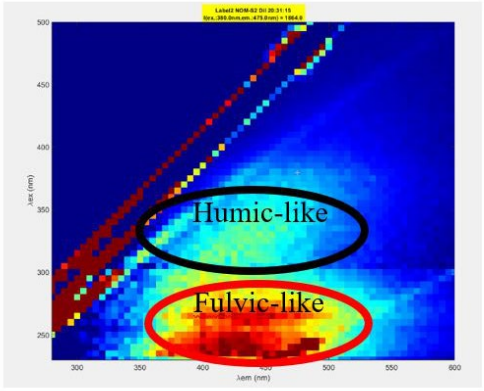

Figure 1: Example of Excitation-Emission Matrix for one of the studied DOM

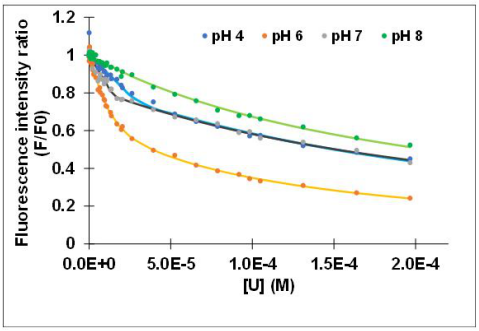

Figure 2: Fluorescence quenching curve obtained for a specific DOM at 4 different pH with their fitted curves. In dot point are the experimental values and the continued line is the corresponding fitted curve