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Redox Speciation of Cerium in the Presence of Natural Organic Matter

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Understanding the interactions between trace metals and natural organic matter (NOM) is crucial due to their significant influence on metal speciation, mobility, toxicity, and bioavailability. Cerium (Ce), a lanthanide element, is of particular interest because of its abundance and unique redox properties, since existing in both Ce(III) and Ce(IV) oxidation states. Various techniques are available to investigate metal-NOM interactions, amongst which differential absorbance spectroscopy (DAS) is considered as a robust method for studying these interactions at environmentally relevant concentrations. However, while DAS has been widely applied to various metals[1], redox-sensitive elements such as cerium and poorly soluble elements such as tetravalent thorium (Th(IV)) and Ce(IV) have not been thoroughly investigated. This gap in the literature limits drastically our understanding of nanoparticle precipitation and redox transformations in the presence of NOM.

This study examines Ce redox speciation and $Ce(OH)_4$ nanoparticle formation at environmentally relevant Ce trace concentrations using DAS. Cerium-NOM interactions were analysed across a pH range between 4 and 8, with praseodymium (Pr(III)) and Th(IV) serving as analogues for Ce(III) and Ce(IV), respectively. Our results indicate that Ce(III) complexation with NOM is most pronounced up to pH 6, where it behaves similarly to Pr^{3+}. At pH 8, $Ce(OH)_4$ nanoparticle formation is favoured, displaying behaviour comparable to Th^{4+}. These findings demonstrate that DAS is an effective technique for elucidating Ce redox speciation and nanoparticle formation under these conditions. Additionally, M-edge X-ray absorption spectroscopy data support these results, confirming Ce redox speciation across the investigated pH range in the presence of NOM.

Overall, this study provides valuable insights into Ce redox behaviour and its interactions with NOM, contributing to a more comprehensive understanding of its environmental fate. These findings can aid in the development of environmental risk assessments and management strategies. In the future, they may also inform predictive models of Ce redox speciation, further enhancing our ability to assess and mitigate the environmental impact of this emerging contaminant.

[1] Chen, Y., Fabbricino, M., Benedetti, M. & Korshin, G. Spectroscopic in situ examination of interactions of rare earth ions with humic substances. *Water Res.* **68**, (2015).