Quantum dot dissolution in photic and aphotic aqueous environments

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Extensive usage of engineered nanoparticles has evoked a need to assess their occurrence and fate within environmental systems upon release to the environment. Quantum dots (QDs) are mixed-metal semiconductor nanocrystals with unique and tunable optical properties that readily leach heavy metal cations in water. QDs distinctive properties including bright fluorescence, wide absorbance range, narrow emission spectrum, and size-dependent emission may lead to unique reactivity when released into the environment, compared to the more homogeneous lattice structures of naturally-occurring bulk metal sulfide or selenide minerals. The goal of this work was to examine dissolution kinetics and mechanisms of QDs relevant to natural surface waters in both photic and aphotic conditions. A systematic approach was used to understand dissolution processes of mercaptopropionic acid capped CdSe/ZnS QDs. QD suspensions were prepared, and release of Zn and Cd cations as well as production of reactive oxygen species under light were monitored over time. The results showed that superoxide and hydrogen peroxide are being produced and are involved in the QDs dissolution. The presence of other compounds such as nitrate, Fe(III), and natural organic matter was observed to improve dissolution kinetics under light. Structural and electronic QD properties (e.g., size, band gap) effect on QD dissolution were investigated, and it was shown that smaller QDs leach core heavy metals faster than larger QDs. In addition, it was observed that shell thickness is the most important structural feature controlling QDs dissolution under light. Results from experiments in the absence of light showed that capping ligand solubility, oxidation by dissolved O2 as well as shell and core solubility in the environment are the important factors in QD dissolution. Results obtained from this study benefits the understanding of fate and transformation of QDs in environmentally relevant low concentrations.