Correlation Between Nitrate and Naturally Occurring Uranium Contamination in Two Major US Aquifers: Potential for Nitrate Driven U Contamination of Groundwater

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Uranium (U) has been identified as a natural contaminant exceeding the US EPA MCL (30 μg l^{-1}) in groundwater throughout the US. This contamination is particularly notable in the High Plains and Central Valley Aquifers, two of the largest aquifers in the US. Alkalinity increases have been traditionally attributed to mobilization of naturally occurring U. However, alkalinity alone cannot explain mobilization of U buried as a reduced U(IV) mineral. As such, oxidation of reduced U(IV) to U(VI) would be required. In addition to molecular oxygen (O_2) one such anthropogenic contaminant, nitrate, is a common oxidant. Nitrate can serve as an abiotic oxidant or as an electron acceptor for microbial oxidation of reduced metals such as U(IV) or Fe(II). Here we use multivariate spatial interpolation of 415,000 groundwater-sampling activities at 138,000 testing locations within High Plains and Central Valley Aquifers to predict nitrate and U contamination. Spatial correlation analysis demonstrated that nitrate is significantly correlated to U exceeding the MCL ($p<0.0001$) in both aquifers. Sediment collected from a shallow aquifer within the High Plains revealed that virtually all U in the sediments existed as U(IV). Although sediments were reducing, oxidized species, nitrate and U(VI), were present in groundwater (>30 mg/L and >30 mg/L). These data suggest potential for U biogeochemical cycling including nitrate driven U(IV) oxidation. Members of bacterial genera capable of anaerobic U(IV) oxidation, *Pseudomonas and Acidovorax*, and U(VI) reduction, *Geobacter*, were identified. Together these data suggest active U biogeochemical cycling. As such nitrate may drive mobilization of naturally occurring U resulting in contamination of groundwater increasing the risk to drinking and irrigation water.