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The effect of gravel size fraction on the distribution coefficients of selected radionuclides

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ABSTRACT

This manuscript addresses the consequences of the common practice of assuming that the gravel fraction of sediments does not participate in sorption reactions and thus sorption quantified by the distribution coefficient (K_d) construct can be estimated from laboratory tests on sediments less than 2 mm size fraction. However, this common assumption can lead to inaccurate estimates of the mobility and sorption affinity of many radionuclides (e.g., Tc, U, and Np) on gravel dominated sediments at the Hanford Site and other locations. Laboratory batch sorption experiments showed that the distribution coefficients measured using only sediment less than 2 mm size fraction and correcting for inert gravel fraction were not in agreement with those obtained from the bulk sediments including gravel (larger than 2 mm size fraction), depending on the radionuclide. The least reactive radionuclide, Tc had K_d values for bulk sediment with negligible deviations from the inert gravel corrected K_d values measured on less than 2 mm size fraction. However, differences between measured K_d values using sediment less than 2 mm size fraction and the K_d values on the bulk sediment were significant for intermediately and strongly reactive radionuclides such as U and Np, especially on the sediment with gravel fractions that contained highly reactive sites. Highly reactive sites in the gravel fraction were attributed to the presence of Fe oxide coatings and/or reactive fracture faces on the gravel surfaces. Gravel correction factors that use the sum of the $K_{d,<2\text{ mm}}$ and $K_{d,>2\text{ mm}}$ values to estimate the K_d for the bulk sediment were found to best describe K_d values for radionuclides on the bulk sediment. Gravel correction factors should not be neglected to predict precisely the sorption capacity of the bulk sediments that contain more than 30% gravel. In addition, more detailed characterization of gravel surfaces should be conducted to identify whether higher reactive sorbents are present in the gravels.

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1. Introduction

Most natural sediments are mixtures of grains of different sizes consisting of varying amounts of the four main particle size classes: gravel, sand, silt, and clay. However, laboratory measurements to characterize the geochemical, physical, and hydrologic properties of sediments and to quantify distribution coefficient (K_d) for contaminant at mineral-solution interfaces are typically performed on gravel-removed size fractions (less than 2 mm). The bulk physical and hydrologic properties of soils and sediments depend strongly on the total mixture of grains, grain shape, and grain packing arrangements. Geochemical process such as adsorption–desorption in the sediments is also affected by the chemical compositions of the fluids within pore spaces surrounding the individual grains as well as the surface reactivity of each solid particle. The fate and transport of dissolved contaminants within pore waters depends on both geochemical reactions (e.g., dissolution/precipitation and adsorption/desorption) and hydrological factors (e.g., diffusive and advective forces) that are controlled by surface area, surface charge, porosity, and packing geometries of the individual grains.

The gravel issue has risen in performance assessment of radionuclide mobility and remediation activity at the Hanford Site because many of the vadose zone and aquifer sediments

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below the Hanford Reservation disposal sites are quite coarse in texture. Because most soil chemistry characterization has been performed on only material less than 2 mm in size, the result effectively leads to the assumption that the gravel fraction is inert (Serne et al., 2002; Um et al., 2007a,b; Williams et al., 2007). Removing the gravel fraction out of the bulk sediment that includes all the size fractions can also disrupt the porosity and grain packing geometries in complicated ways. In addition, because gravel or larger particles are removed prior to measurement of common geochemical parameters such as cation exchange capacity (CEC) and sorption distribution coefficient (K_d), the results of most geochemical characterization on sediment less than 2 mm size fraction are not directly representative of the natural field conditions. Often Fe and Al oxide coatings, which affect sorption of metal contaminants, are found in the gravel fraction of aquifer sediments (Coston et al., 1995; Penn et al., 2001; Davis et al., 2004; Kent and Fox, 2004; Um et al., 2007a,b). Ignoring

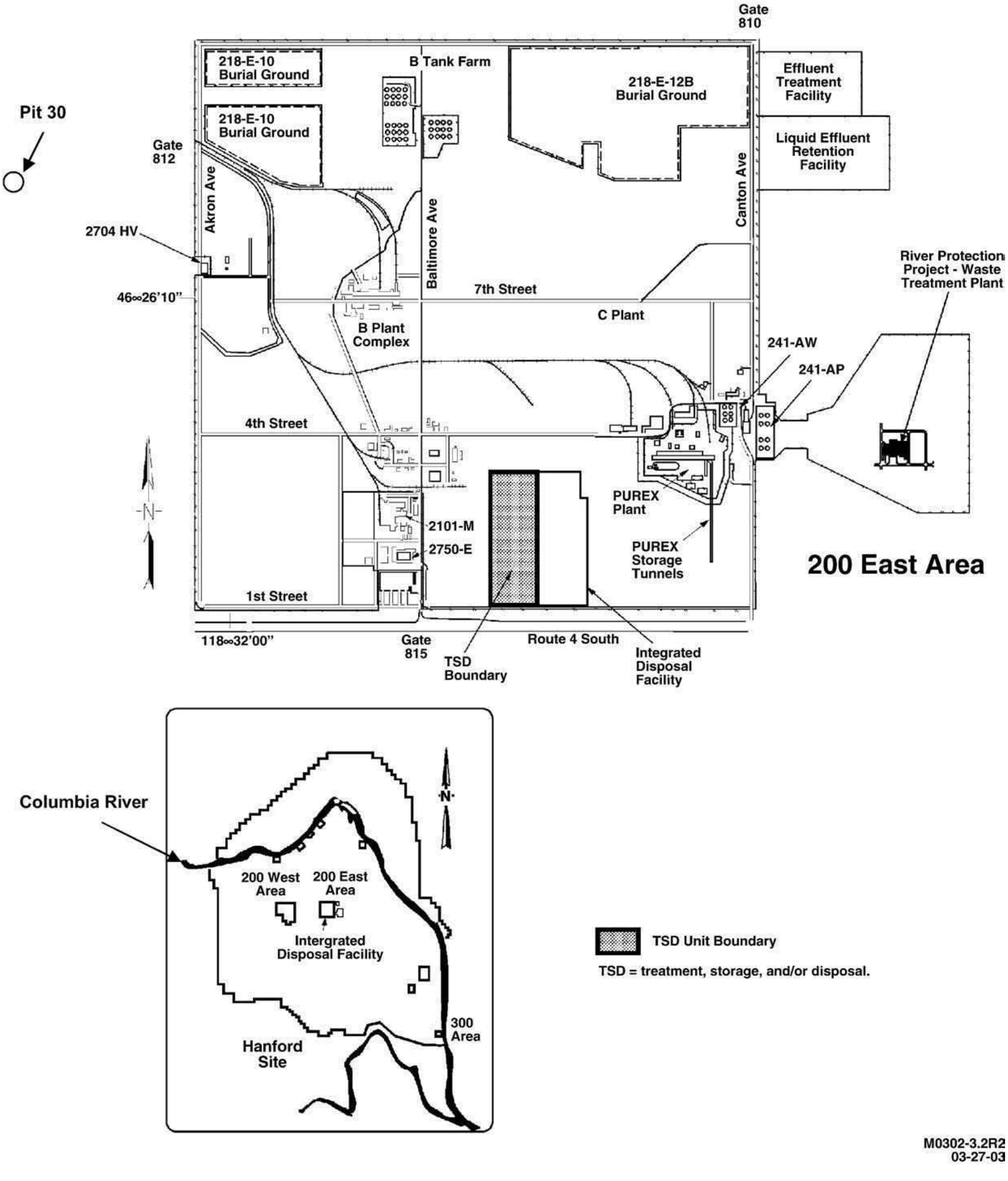

Fig. 1. Integrated disposal facility (IDF) location on the Hanford Site.

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