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Analysis of trace neptunium in the vicinity of underground nuclear tests at the Nevada National Security Site

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ABSTRACT

A high sensitivity analytical method for ^{237}Np analysis was developed and applied to groundwater samples from the Nevada National Security Site (NNSS) using short-lived ^{239}Np as a yield tracer and HR magnetic sector ICP-MS. The ^{237}Np concentrations in the vicinity of the Almendro, Cambric, Dalhart, Cheshire, and Chancellor underground nuclear test locations range from $<4 \times 10^{-4}$ to 2.6 mBq/L (6×10^{-17}–4.2×10^{-13} mol/L). All measured ^{237}Np concentrations are well below the drinking water maximum contaminant level for alpha emitters identified by the U.S. EPA (560 mBq/L). Nevertheless, ^{237}Np remains an important indicator for radionuclide transport rates at the NNSS. Retardation factor ratios were used to compare the mobility of ^{237}Np to that of other radionuclides. The results suggest that ^{237}Np is less mobile than tritium and other non-sorbing radionuclides (^{14}C, ^{36}Cl, ^{99}Tc and ^{129}I) as expected. Surprisingly, ^{237}Np and plutonium (^{239,240}Pu) retardation factors are very similar. It is possible that Np(IV) exists under mildly reducing groundwater conditions and exhibits a retardation behavior that is comparable to Pu(IV). Independent of the underlying process, ^{237}Np is migrating downgradient from NNSS underground nuclear tests at very low but measureable concentrations.

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1. Introduction

1.1. Environmental relevance of ^{237}Np

The alpha-particle emitter ^{237}Np is the most relevant Np isotope with regards to environmental contamination. The majority of ^{237}Np in the global inventory is produced via neutron irradiation of ^{235}U and ^{238}U in nuclear fuel (Thakur and Mulholland, 2012), predominantly as a result of successive neutron capture on both ^{235}U and ^{238}U followed by β-decay:

$$^{235}\mathrm{U}(n,\gamma)\ ^{236}\mathrm{U}(n,\gamma)\ ^{237}\mathrm{U}(\beta^-) \rightarrow\ ^{237}\mathrm{Np}$$

$$^{238}\mathrm{U}(n,2n)\ ^{237}\mathrm{U}(\beta^-) \rightarrow\ ^{237}\mathrm{Np}$$

With respect to the nuclear fuel cycle, nearly all ^{237}Np produced from nuclear fuel is destined to be incorporated into high level waste (HLW). This is primarily because no practical application of ^{237}Np exists, while Pu and U are recycled through commercial reprocessing of spent nuclear fuel. As a result, it is anticipated that ^{237}Np will be a major contributor to the total radiation in spent nuclear fuel and the associated HLW after 10,000 years. Due to its long half-life (2.14×10^6 years) and relatively high predicted mobility under aerobic conditions, ^{237}Np has also been identified as a key long-term dose contributor in certain nuclear waste repository scenarios (DOE (US Department of Energy), 2002). As a result, identifying the factors affecting ^{237}Np transport in the environment is essential for helping to design a safe, long-term nuclear waste repository.

1.2. ^{237}Np at the Nevada National Security Site (NNSS)

The Nevada National Security Site (NNSS, formerly Nevada Test Site (NTS)) was the primary location of nuclear weapons testing for the United States. Between 1951 and 1992, over 800 underground nuclear tests were detonated at the NNSS. Approximately one-third of these tests was located below the water table (DOE/NV, 2000), which has led to significant radiologic groundwater contamination. The radioactive material deposited after an underground nuclear test, which is referred to as the radiologic source term, consists of tritium, fission products, actinides, and activation products. At the NNSS, the total activity deposited in the subsurface is approximately 4.9×10^9 GBq (1.3×10^8 Ci at 1992) and consists of as many

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as 43 radioactive isotopes (Bowen et al., 2001; Smith et al., 2003) with half-lives greater than 10 years (with the exception of ^{154}Eu with a half-life of 8.6 years). Predicting the transport behavior and ultimately the environmental risk of this radiologic source term is the subject of active investigation by the Environmental Management program of the Department of Energy (DOE/CF, 2013). The dominant mobile (non-sorbing) radiologic contaminants in groundwater at the NNSS are expected to be ^{3}H, ^{14}C, ^{36}Cl, ^{99}Tc, and ^{129}I (Hu et al., 2008). Less mobile radiologic contaminants include ^{137}Cs, ^{90}Sr, Pu and ^{237}Np. However, despite the mobility perspectives, Pu was found to migrate from the original detonation site (Kersting, 2013; Kersting et al., 1999). The presence of the underground radiological source term at the NNSS provides a unique opportunity to study the migration behavior of a large number of radionuclides that are relevant for nuclear waste repository performance assessments and for understanding the evolution of radiologic plumes at nuclear accident sites.

The main sources of ^{237}Np in underground nuclear tests are (1) radiochemical tracer and (2) the decay of ^{237}U (Bowen et al., 2001; Smith et al., 2003). Some additional ingrowth of ^{237}Np will occur from the ^{241}Pu/^{241}Am decay chain. As of 1992, ^{237}Np contributed only 1.8 TBq (3×10^2 moles) versus 3.1×10^4 TBq from plutonium (1.2×10^4 moles) to the total radiologic source term (4.9×10^6 TBq), which is dominated by ^{3}H (4.7×10^6 TBq, 4.3×10^3 moles) (Bowen et al., 2001; Smith et al., 2003). However, it has been considered a relevant actinide in environmental risk assessments due to its long half-life, toxicity and mobility in oxidizing environments.

To date, a very limited number of ^{237}Np measurements has been reported for NNSS groundwater. Most of these reported values have been near the method detection limits (e.g. (Finnegan and Thompson, 2002)), which limits quantitative interpretation of Np transport behavior. For example, in 1965 the Cambric test (Fig. 1) was detonated in alluvium below the water table. Subsequently, a complex radionuclide migration experiment was conducted in which groundwater was pumped from a nearby pumping well (RNM-2S) while radionuclide migration from the Cambric test was monitored at a drillback well (RNM-1). Groundwater pumped from RNM-2S was released into a surface drainage that allowed radiologically contaminated water to percolate through the vadose zone and reach a nearby monitoring well (UE-5n). ^{237}Np concentrations were previously reported to be the same at all three wells: 1.9 mBq/L (3×10^{-13} mol/L) with a method detection limit of 0.06 mBq/L (10^{-14} mol/L) (Finnegan and Thompson, 2002). In contrast, recent Cambric reactive transport simulations predicted ~0.6 mBq/L (10^{-13} mol/L) ^{237}Np at the drillback well (RNM-1), ~0.006 mBq/L (10^{-15} mol/L) ^{237}Np at the pumping well (RNM-2S), and little or no ^{237}Np at the monitoring well (UE-5n) (Carle et al., 2005). It is not clear whether the discrepancy between measured and predicted values can be attributed to measurement uncertainties or inaccurate model predictions. Validation of these modeling simulations requires an improved resolution of ^{237}Np concentrations in NNSS groundwater wells.

1.3. Analysis of ^{237}Np in environmental samples

Major challenges in the analysis of ^{237}Np in environmental samples are its ultra-low concentrations, the lack of suitable yield tracers and the need to separate ^{237}Np from interfering constituents in environmental samples. For instance, the quantification of low-level ^{237}Np in NNSS groundwater is complicated by the presence of approximately seven orders of magnitude higher natural uranium concentrations (μg/L levels), which interferes with ^{237}Np analysis by inductively coupled plasma mass spectrometry (ICP-MS). Radionuclide measurement techniques for environmental samples, including both radiometric and mass spectrometric techniques, have been recently critically reviewed by several

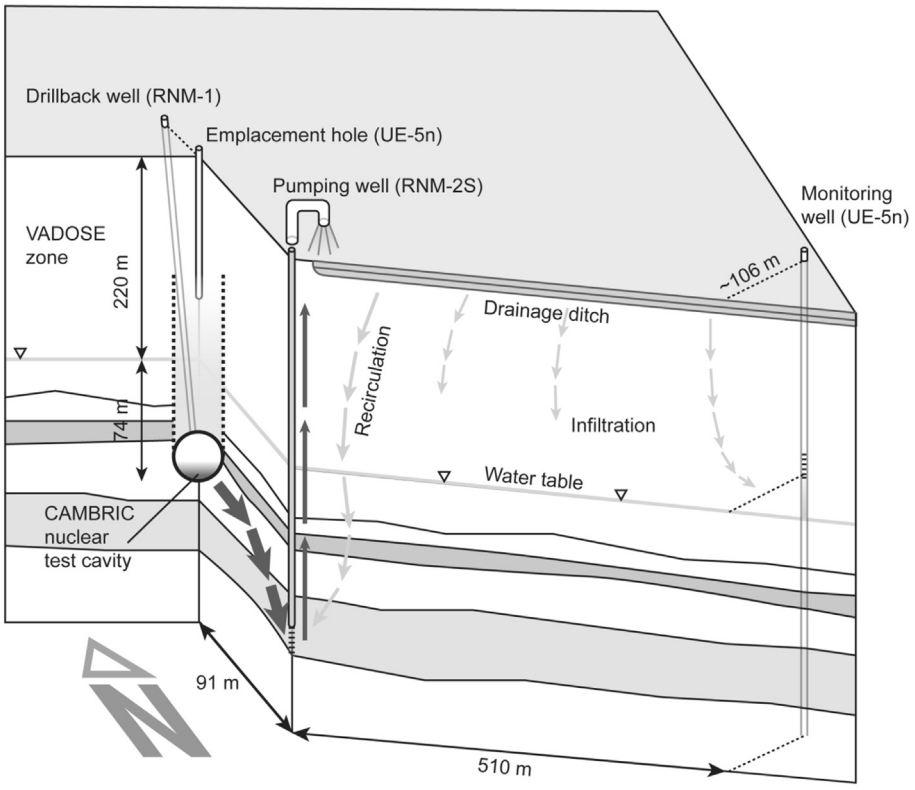

Fig. 1. A view of the Cambric site with associated groundwater sampling wells (RNM-1, RNM-2S, and UE-5n) and their locations. From Carle et al. (2005).

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