Available online at www.sciencedirect.com
SCIENCE DIRECT®

Journal of Environmental Radioactivity 89 (2006) 61–80

JOURNAL OF
ENVIRONMENTAL
RADIOACTIVITY
www.elsevier.com/locate/jenvrad

A comparison of the soil migration and plant uptake of radioactive chlorine and iodine from contaminated groundwater

D.J. Ashworth*, G. Shaw[1]

Department of Environmental Science and Technology, Imperial College London,
Silwood Park, Ascot, Berkshire SL5 7PY, UK

Received 14 September 2005; received in revised form 17 March 2006; accepted 28 March 2006

Abstract

A 6-month soil column experiment was conducted to compare the upward migration and plant uptake of radiochlorine and radioiodine from shallow, near-surface contaminated water tables. Both fixed and fluctuating water tables were studied. After 6 months, ^{36}Cl activity concentrations were relatively uniform throughout the soil profile apart from an accumulation at the soil surface, which was especially marked under a fluctuating water table scenario. In contrast, ^{125}I (a surrogate for ^{129}I) tended to accumulate at the boundary between the anoxic conditions at the base of the column and the oxic conditions above, due to its redox-dependent sorption behaviour. The uptake of ^{36}Cl by perennial ryegrass was much greater than that of ^{125}I due to its greater migration into the rooting zone and its ready availability in soil solution. In the context of radioactive waste disposal, where these radionuclides may potentially be released into groundwater, ^{36}Cl would be expected to present a greater potential for contamination of the biosphere than ^{129}I.

Keywords: ^{36}Cl; ^{129}I; Radioactive waste disposal; Iodine speciation; Redox potential; Organochlorine

---

* Corresponding author. USDA-ARS, George E Brown Salinity Laboratory, 450 W. Big Springs Road, Riverside, CA 92507, USA.

E-mail address: dashworth@ussl.ars.usda.gov (D.J. Ashworth).

[1] Current address: Division of Agricultural & Environmental Sciences, University of Nottingham, University Park, Nottingham NG7 2RD, UK.

0265-931X/$ - see front matter © 2006 Elsevier Ltd. All rights reserved.
doi:10.1016/j.jenvrad.2006.03.006

1. Introduction

The radionuclides ^{36}Cl and ^{129}I are, together with other radionuclides, potential components of intermediate and high level radioactive waste. Their primary source in such waste is the operation of nuclear power plants in which they are formed within the reactor core. The ^{36}Cl is formed due to neutron capture by stable ^{35}Cl, which may be present at trace levels in the core materials. The ^{129}I is produced by the fission of uranium atoms. One potential option for the disposal of intermediate level radioactive waste is burial within a deep geological repository. Such repositories would be engineered in such a way as to try and minimise the release of radionuclides into the host rock and hence to the overlying biosphere. Nevertheless, it has been deemed prudent by radioactive waste disposal agencies to assess the overall risk associated with the leakage of radionuclides from a repository site. The radionuclides of primary interest in this regard are those that have a long physical half-life and a potentially high degree of environmental mobility. Both ^{36}Cl and ^{129}I, which are likely to be predominantly found in anionic form in rock at depth and have physical (radioactive) half-lives of 3.01×10^5 and 1.57×10^6 years, respectively, fulfil these criteria.

Part of such a risk assessment involves quantifying the behaviour of radionuclides within the biosphere, including their potential for migration through soils and their subsequent transfer from soil to plants. Experimental studies are, therefore, required to elucidate the processes governing radionuclide behaviour within the biosphere. In the context of possible leakage of radionuclides from a waste repository, in which case contamination of the groundwater would result, the upward migration of radionuclides towards the soil surface and the rooting zone of vegetation is one of the pathways of interest. In this paper, an experimental study of the upward soil migration and plant uptake of ^{36}Cl and ^{125}I (as a surrogate for ^{129}I) from a contaminated water table is described. The primary objectives of the work were to determine (a) the extent of upward soil migration of these radionuclides in relation to hydrological (water table movement and evapotranspiration flux) and chemical (sorption) processes, and (b) the degree of plant uptake of these radionuclides in relation to radionuclide and root distributions within the soil.

Chlorine behaviour in soils is considered to be dominated by its presence as the inorganic chloride form (Brady and Weil, 1996). This conservative (non-sorbed) form is often assumed to follow hydrological flows almost quantitatively and is thus routinely used as a tracer for the movement of water through, for example, soil columns (Ashworth and Alloway, 2004), natural soils in the field (Newman et al., 1997) and aquifers (Murad and Krishnamurthy, 2004). Furthermore, ^{36}Cl was shown by Ogard et al. (1988) to break through to the surface environment following underground nuclear testing slightly before tritiated water, suggesting that the migration of radiochlorine may be enhanced due to anion exclusion (Thomas and Swoboda, 1970). This high affinity for the water phase of soils results in the solid-solution partition coefficient (K_d value) of chlorine generally being assumed to be zero. In addition, it also renders chlorine biologically available. For example, Shaw et al. (2004) calculated ^{36}Cl transfer factors for winter wheat in field lysimeters which ranged from <1 to around 6400, with a mean of around 700. These extremely high transfer factors indicated that up to 40% of the ^{36}Cl added to the soil was taken up by the winter wheat crop in a single growing season. Therefore ^{36}Cl is expected to be one of the most mobile and biologically available of the radionuclides that are important in the context of radioactive waste disposal.

Chemical species of chlorine in soils, other than chloride, are not often reported. However, a small but significant body of work, initiated by Asplund and Grimvall (1991), suggests a role of natural organic matter in the formation of organo-chlorine species within soils. Oberg (1998) specifies two processes by which the formation of chlorinated soil organic matter may come