Cl-36 Transfer to Ryegrass and Consequences for Environmental Modeling

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INTRODUCTION

Chlorine-36 is a long-lived and highly mobile radionuclide that has not historically received much attention. In recent years that has begun to change and there is particular current interest in the transport and fate of Cl-36 in the environment. In September of 2006 ANDRA hosted an International Forum on Chlorine-36 in the Biosphere where models indicating that Chlorine 36 will be a significant contributor to long-term potential doses arising from radioactive waste disposal were presented. A recent report of the International Union of Radioecology's Radioecology and Waste Task Force identified the need for more research, stating in part: "More experiments for the determination of Cl-36 soil to plant transfer should be performed and combined with studies of stable Cl behaviour. In particular, direct measurement of uptake chlorine fluxes by forest vegetation and reliable estimations of budgets are needed (IUR, 2006)."

The data that does exist varies widely, in one study concentration ratios from less than 1 to more than 6400 were calculated, with 40% of the total chlorine 36 inventory concentrated in plant tissue after one growing season (Shaw et al., 2004). Other investigators have recorded similar results, showing that Cl-36 is both highly mobile, and highly bioavailable (Ashworth and Shaw, 2006; Coughtrey et al., 1983; Kashparov et al., 2007; Shepard et al., 1993). Chloride has traditionally been considered to be chemically inert and to behave conservatively in soil with very low sorption values, but recent research suggests that chloride participates in a complex biogeochemical cycle that forms and mineralizes organically bound chlorine (Lee et al., 2001; Oberg and Sanden, 2005).

These experiments were designed to increase the body of data on chloride uptake in plants. Rye grass (*Lolium perenne*) was grown in 5 lysimeters. Cl-36 in an inorganic chloride form was introduced to the system through a contaminated water table, and over a period of months, data was taken on the movement of the Cl-36 through the soil and its uptake in ryegrass.

MATERIALS AND METHODS

Lysimeter construction

In an experimental design after that of Ashworth and Shaw (2005), each lysimeter consisted primarily of a 60 cm length of PVC tubing with a 15 cm diameter. To make later analysis easier, the PVC was cut lengthwise then resealed with silicon sealant and bound with jubilee clips. The columns were sealed at one end and a small hole drilled 2.5 cm from the sealed base of the column. This hole was fitted with a tube leading to the plastic base of a 1 L graduated cylinder serving as an exterior reservoir that maintained the water table at the desired height. The bottom 5 cm of the column were packed with polythene beads, above which was placed a mesh filter, the remainder was filled with soil. The soil used in these experiments was obtained from a local

landscaping company and was determined by the Oregon State University Central Analytical Laboratory to be a sandy loam with a particle size distribution of 80% sand, 11.3% silt, and 8.8% clay. The soil has a pH of 7.2, a stable chloride content of 3.5 ppm, and an organic matter content of 2.27% based on a loss on ignition test. Five columns were used in these experiments; three of the five columns were fitted with three Rhizon soil pore water samplers placed at 20 cm intervals.

Experimental Setup

Five lysimeters were deployed in a small heated greenhouse keeping an18 hours on, 6 hours off lighting schedule. For two weeks before the introduction of Cl-36 a water table was established 20 cm from the base of the columns. The Chlorine-36 used in these experiments was introduced in a chloride form at a concentration of 20kBq/liter. Planting and the introduction of Cl-36 both occurred on day 1 of the experiments, at which time the soil surface was watered with DI water to aid in germination. After this preliminary watering, all water was introduced via the exterior reservoir.

Analytical procedures

Soil pore water

Soil pore water was sampled on a weekly basis at three levels of the three lysimeters.
1 ml aliquots of the weekly samples were added to 6 ml of Optiphase HiSafe II LSC
fluid.

Plant tissue

Plant samples were weighed, then dried and maintained at 60 ºC. Before digestion samples were ground and homogenized. Two methods of digestion were employed. In the first digestion type, 200 mg of dried plant matter was digested under reflux with 5ml concentrated nitric acid for 4 hours, 1 hour at 90 ºC after which the temperature was raised to 120 ºC. 100 uL aliquots of the resulting solution were added to 6 ml of Optiphase HiSafe II LSC fluid for counting. (Ashworth and Shaw, 2006; Ashworth personal communication, 2008) The alternate digestion method employed used Soluene 350. 100 mg of dried plant matter was placed in a 20 ml LSC vial and combined with 6 ml of Soluene 350. The vial was then kept overnight in an oven at 60 ºC and allowed to cool before 14 ml of Hionic-Fluor LSC fluid was added for analysis.

Soil

Soil samples were tested in 5 g samples. Samples were placed in a 50 mL plastic centrifuge tubes. 35mL of deionized water was added and the sample was shaken for two hours. The samples were then centrifuged for 15 minutes before aliquots were removed for LSC. By completing a set of additional extractions using NaOH and HCl as extractants instead of DI water, knowledge of the extent of Cl-36 sorption to the soil can be gleaned.

RESULTS

This abstract describes an ongoing series of experiments and the results discussed in this

extended abstract are preliminary. For this reason a complete inventory of each lysimeter's chlorine-36 contents is not yet available. A small subset of the available data is presented here to support the conclusion that the conventional wisdom is correct, and Cl-36 is both mobile and highly bioavailable in ecological systems.

Figure 1 is a graph of the soil pore water activities at 10, 30, and 50 cm above the base of the lysimeter. The levels initially spiked at the lowest sampling location in each lysimeter after planting. Cl-36 introduction occurred on day 1 of the experiments, and these spikes are attributed to a buildup of Cl-36 that continues until the ryegrass roots reach the water table, after which values fall as the root system appears to take up the majority of available Cl-36. A final, destructive, analysis of the lysimeters will allow a complete mass balance inventory of the Cl-36 introduced into each lysimeter, with the fraction in soil, and plant tissue, and allow for the calculation of the sorption values.

Initial harvests of the aboveground portions of ryegrass have yielded dry weight concentrations of 11 to 66 Bq/g. These numbers are expected to rise as uptake continues and root systems are examined after destructive analysis of the lysimeters.

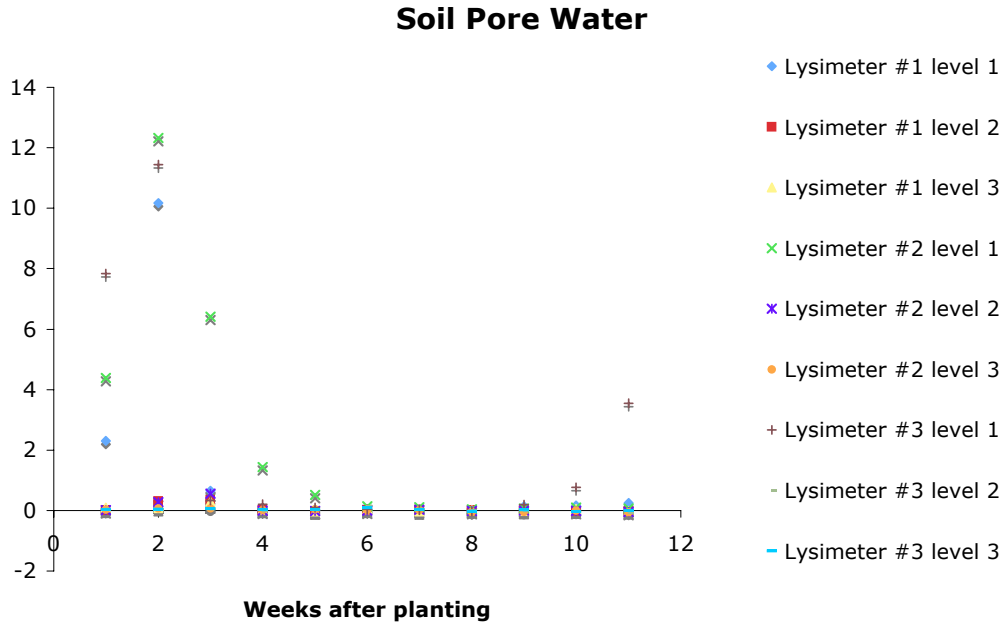

Figure 1. Three months worth of soil pore water data from three lysimeters.

CONCLUSIONS

Chlorine-36 in a chloride form shows a high degree of bioavailability. The soil pore water data indicates that as root systems become established, they are capable of uptaking virtually all of the Cl-36 within their reach. The extent of uptake is affected by the presence and concentration of stable chloride, but the potential for uptake marks Cl-36 as an ideal candidate for potential phytoremediation of contamination.

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