

Cl-36 transfer to ryegrass and consequences for environmental modeling

D. Bytwerk and K. Higley

Department of Nuclear Engineering & Radiation Health Physics, Oregon State University,
Corvallis, OR 97331, USA

Abstract. There has been recent interest in the transport and fate of ^{36}Cl in the environment. The IUR task force on Radioecology and Waste has identified the need for further uptake data. Reported here are the results of a 6-month study of the uptake in ryegrass of ^{36}Cl in five lysimeters. Lysimeters were deployed in a greenhouse from November 2007 to May 2008. Except for an initial watering, exterior reservoirs connected to the base of the lysimeters maintained the water tables at a constant level with a solution containing ^{36}Cl in a chloride form. A destructive analysis of the lysimeters showed that between 75 and 80% of ^{36}Cl added to the system during the course of the experiments was taken up into the plant tissue. The quantities of ^{36}Cl found in weekly soil tests and a simple mass balance provide supporting evidence for the significant bioavailability of ^{36}Cl in a chloride form.

1. BACKGROUND

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 ^{36}Cl in the environment. In September of 2006 ANDRA hosted an International Forum on Chlorine-36 in the Biosphere where models indicating that ^{36}Cl 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 ^{36}Cl 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 [6]."

Chlorine is an essential micronutrient for plant growth, but it is required in extremely small amounts. There are significant environmental reserves of chloride and so chlorine is almost never a limiting factor in plant growth. As a result chlorine uptake doesn't scale as reliably with mass as macronutrients such as nitrogen or phosphorous. This coupled with chloride's mobility in soil and propensity for uptake means that plant concentrations can vary widely depending on environmental conditions.

The data that do exist vary widely but consistently demonstrate the bioavailability of ^{36}Cl . Shaw, Wadey, and Bell [9] report concentration ratios from less than 1 to more than 6400, with 40% of the total ^{36}Cl inventory concentrated in plant tissue after one growing season. Colle et al. [3] reported even higher values, "60% of the contamination was extracted from the soils by the plants after one vegetative period." Other investigators have recorded similar results, showing that ^{36}Cl is both highly mobile, and highly bioavailable [2, 4, 7, 11]. 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 [8, 9].

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

2. MATERIALS AND METHODS

2.1 Lysimeter construction

In an experimental design after that of Ashworth and Shaw [1], 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. Soil was packed into the lysimeter in 10 cm sections in order to compact the soil to a density of approximately 1.1 g/cm³. 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. These were installed by drilling small holes at 10, 30, and 50 cm, placing the soil moisture probes in place and sealing the joint with silicon sealant.

2.2 Experimental setup

Five lysimeters were deployed in a small heated greenhouse keeping an 18 hours on, 6 hours off, lighting schedule. For two weeks before the introduction of ³⁶Cl a water table was established 20 cm from the base of the columns. The ³⁶Cl used in these experiments was introduced in a chloride form at a concentration of 20 kBq/liter. Planting and the introduction of ³⁶Cl 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.

2.3 Analytical procedures

2.3.1 Soil pore water

Soil pore water was sampled on a weekly basis at three levels of the three lysimeters. To take the weekly samples a syringe was attached to one end and used to provide a vacuum pulling soil pore water into the syringe. 1 ml aliquots of the weekly samples were added to 6 ml of Optiphase HiSafe II LSC fluid for analysis.

2.3.2 Plant tissue

Plant samples were weighed, then dried and maintained at 60 °C. Samples were ground before digestion. For each plant sample 150 mg of dried plant matter was added to 5 ml concentrated nitric acid. It was heated under reflux for four hours, one hour at 90 °C after which the temperature was raised to 120 °C for the remaining three hours. 50 uL aliquots of the resulting solution were added to 6 ml of Optiphase HiSafe II LSC fluid for counting. This method was adapted from one used by Ashworth and Shaw [2]. Blank and spiked sample digestions were done to measure the effects of quench and the efficiency of the digestion. An extraction efficiency of 41% was calculated, similar to the 45% recorded by Ashworth and Shaw (personal communication) [2].

2.3.3 Root mass determination and wastewater sampling

Lysimeters were cut into 5 cm sections and sieved through a #35 mesh. Roots were separated from the remaining rock and soil with tweezers and rinsed until they no longer clouded water in which they were placed. Total root masses for each 5 cm section were measured after drying them to a constant weight. The water used to wash the soil through the sieve was retained and used to take 1 ml aliquots which were added to 6 ml of Optiphase HiSafe II LSC fluid for counting. Chloride is commonly considered to have a solid-solution partition coefficient k_d of 0 [2, 5]. Although Lee et al. [8] have reported different results, for the purposes of a conducting a simple mass balance the wastewater will be assumed to contain all of the ^{36}Cl that had been associated with the soil.

3. RESULTS

A significant amount of data was collected over the course of the experiment. A small subset of the available data is presented here to support the conclusion that the conventional wisdom is correct, and ^{36}Cl is both mobile and highly bioavailable in ecological systems. Data is presented on the activity concentration in dried plants in all lysimeters, soil pore water data for three lysimeters, and a simple mass balance will be presented with a root depth profile for lysimeters #1 and #2.

3.1 Soil pore water data

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. Chlorine-36 introduction occurred on day 1 of the experiments, and these spikes are attributed to a buildup of ^{36}Cl 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 ^{36}Cl . The spike towards the end of the experiments is attributed to a decrease in uptake as the plants ran out of resources in the nutrient poor soil used in these experiments and growth rates slowed dramatically. The ^{36}Cl values in the samplers higher in the lysimeters never recorded significant levels of ^{36}Cl .

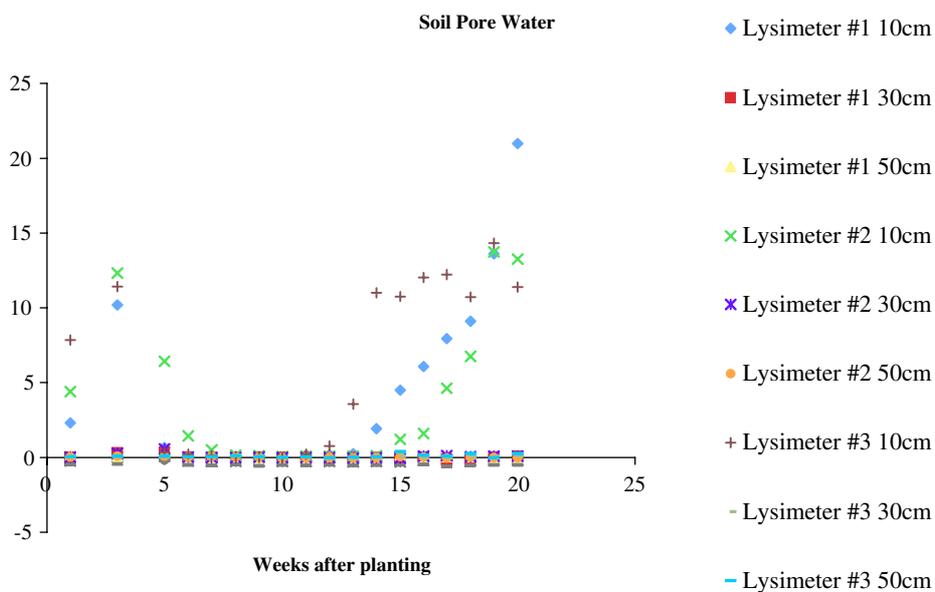


Figure 1. Three months worth of soil pore water data from three lysimeters.

3.2 Plant activity concentrations

Plant activity determination was done using dried plant tissue harvested from the aboveground portion of the grasses. A difference in chloride content in the aboveground and belowground portions of the plant could be a potential source of error. Values for the first three lysimeters are significantly larger than those of the latter two. One cause for this is that the first three were instrumented with Rhizon soil moisture probes, and every week 10 ml of water was pulled out from the system at various heights, this led to more ^{36}Cl being dispensed to these three lysimeters to maintain the same constant water level.

Table 1. Plant activity concentration data.

	Dry weight activity concentration (Bq/g)	Uncertainty (Bq/g)
Lysimeter #1	8065.9	± 217.8
Lysimeter #2	8755.9	± 296.0
Lysimeter #3	6404.1	± 253.6
Lysimeter #4	5428.5	± 233.4
Lysimeter #5	4504.5	± 213.1

3.3 Root depth distribution

Figure 2 is included for some perspective on how far the roots penetrate. Since the roots easily reach the depth of the contaminated water table, the ^{36}Cl can be uptaken without relying on evapotranspiration or other water fluxes to lift the ^{36}Cl to the roots.

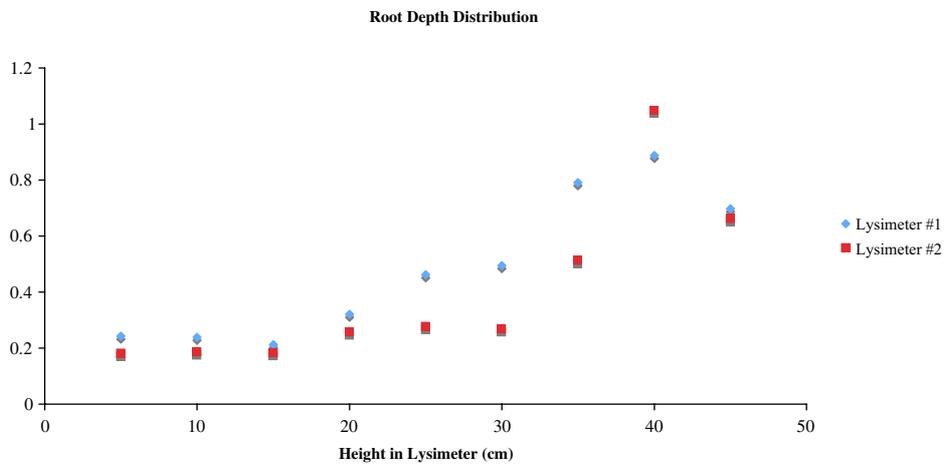


Figure 2. Root mass by height in lysimeters #1 and #2.

3.4 Total uptake in plant material and a simple mass balance

Lysimeters #1 and #2 had enough data for a simple mass balance analysis. Table 2 contains the data from each lysimeter. Dividing the Plant Material Activity by the total activity input to the system allows for the calculation of the total contamination extracted from the soils by the plants after one vegetative period. Lysimeter #1's plant material contained 81% of the system's activity. Lysimeter #2's plant material contained 75% of that system's total activity. The "Missing" radioactivity could be associated with the soil in an organic form, it could be a result of incomplete removal of ^{36}Cl from the soil by the sieving

process, or it could be a result of inhomogeneity in the plant tissue activity concentration, which was used to establish total plant material activity.

Table 2. A simple mass balance.

	Lysimeter #1 Activity	Lysimeter #2
Total System Input	100%	100%
Plant Material Activity	81.3%	75.5%
Wastewater Activity	16.1%	16.9%
“Missing” Radioactivity: Total System Input – (Plant Material Activity + Wastewater Activity)	2.5%	7.6%

4. CONCLUSIONS

Chlorine 36 in a chloride form shows a high degree of bioavailability. The total uptake results presented here are higher than other reported values, but in the same range.

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. This result is confirmed in the plant activity concentration data and the rough mass balance presented above. The extent of uptake is affected by the presence and concentration of stable chloride and other nutrients, but the affinity for uptake marks Cl-36 as an ideal candidate for potential phytoremediation of contamination.

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