AN ABSTRACT OF THE THESIS OF

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Abstract approved:

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Greenhouse studies were conducted to determine interception, absorption, and translocation values for foliar applied ^{36}Cl. Foliar interception and uptake of contaminated irrigation water by crops is a major pathway for the transport of radionuclides to human beings in scenarios relevant to the waste disposal community. Performance assessments of many repositories that predict doses to people from long-term geological disposal of radioactive waste are equipped to consider foliar interception and uptake by crops on an element and crop specific basis in their predictions, but crop and element specific data does not exist in the literature to justify the choice of parameter values in these models. ^{36}Cl is among the isotopes for which there is a lack of data and has recently been predicted to be among the largest contributors to dose at the time of peak dose from many repositories. Reported here are ^{36}Cl foliar interception, absorption, and translocation parameters for three crops: a root vegetable -radishes, a fruit - beans, and a grain - wheat.

Foliar Interception and Uptake of ^{36}Cl by Crops

by
David Paul Bytwerk

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APPROVED:

Major Professor, representing Radiation Health Physics

Head of the Department of Nuclear Engineering and Radiation Health Physics

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Dean of the Graduate School

I understand that my dissertation will become part of the permanent collection of Oregon State University libraries. My signature below authorizes release of my dissertation to any reader upon request.

David Paul Bytwerk, Author

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1 Introduction: The Problem of Waste

Common to most definitions of waste both in language and in law is the desire of its owners to dispossess themselves of it (Pongrácz 2002); waste is not an asset to its owners but rather a liability that requires some expenditure to be removed from the books. Unfortunately waste is ubiquitous in modern society; waste management and disposal make up significant portions of the total work involved in many activities (Milnes 1985). The problem facing society is what to do with things no one wants: the most common solution has been to take the waste and bury it in places engineered to contain it; in the case of municipal solid waste these places are landfills. If the waste in question is innocuous its disposal is relatively straightforward, though even innocuous waste must be disposed of in a regulated fashion given the scale at which it is produced. The disposal of wastes that pose particular hazards to human health presents more challenging technical and regulatory problems, and hazardous waste disposal is rightfully held to higher standards than other wastes. Radioactive waste is a subset of hazardous waste that poses unique challenges. The precise definitions and classifications of radioactive waste vary from jurisdiction to jurisdiction, but a pithy definition from Wiltshire (1993) is: “the radioactive by-products of nuclear weapons production, nuclear power generation, and other uses of nuclear materials. They range from highly radioactive discarded nuclear fuel to slightly radioactive used clothing.” What to do with radioactive waste is an issue facing every country using radioactive materials; countries with significant nuclear energy production possess considerable

volumes of highly radioactive spent fuel and even countries without commercial nuclear power industries will accumulate radioactive waste due to material used in medical diagnostics and therapy, or through use of radioisotopes in research and industry.

A chief way in which radioactive wastes differ from other hazardous wastes is in the longevity of the risk they represent. Milnes (1985) writes: “A rule of thumb of the early nuclear industry was that the confinement or isolation time of radwaste should be sequestered for at least 10 times the half-life of the longest living of the dominant isotopes.” The passage of 10 half-lives ensures, assuming no ingrowth from a parent nuclide, that the remaining radioactivity of that nuclide is less than a thousandth of the initial value. By this back of the envelope method, spent nuclear fuel where ^{239}Pu with a half-life of 24,110 years is a dominant nuclide would require 241,100 years of sequestration, which is within the range of regulatory compliance periods currently debated.

Various solutions to the problem of radioactive waste have been proposed: disposing of the wastes in rockets to space (Rice et al. 1982), disposal of the waste in repositories in the sediments of the deep seabed (Tang and Saling 1990), and transmutation of the waste into less hazardous forms using specialized reactors such as the sodium-cooled fast reactor (Lineberry and Allen 2002). There have even been more radical, and ill-judged, proposals to: “dilute it to a low radiation level and sprinkle it over the ocean -

or even over America after hormesis is better understood" (Robinson 1997). However "there is a worldwide consensus that deep geological disposal… is the best option for disposing of high-level radioactive waste" (NRC 1990). Reduced to its simplest form the idea is to bury the waste at such a place and in such a way that it will never pose unacceptable risks to human health. In addition to the United States, the following countries have programs for the development of geological repositories: Belgium, Canada, China, Finland, France, Germany, Japan, Russia, Spain, Sweden, Switzerland, and the United Kingdom (DOE 2001). These countries face many of the same challenges that the United States does in developing repository programs.

Though there is a consensus that geological disposal of waste is the best available option, isolating waste from the human biosphere for the hundreds of thousands of years required for decay is a singular technical challenge. Geological disposal has been considered since at least 1957 when the National Academy of Sciences published "The Disposal of Radioactive Waste on Land" which found: "Wastes may be disposed of safely at many sites in the United States…the research to ascertain feasibility of disposal has for the most part not yet been done. Disposal in cavities mined in salt beds and domes is suggested as the possibility promising the most practical immediate solution to the problem" (NRC 1957). The essential outline of the geological repository was contained in the 1957 report; the repository, located hundreds of feet below ground, would isolate waste in a stable geological formation where a combination of

geology and engineered systems would prevent release of radionuclides to the environment in levels hazardous to people.

In the United States weapons related waste has been collecting for over 60 years while spent commercial nuclear fuel has been accumulating for over 50 years. The Nuclear Waste Policy Act of 1982 states "the Federal Government has the responsibility to provide for the permanent disposal of high-level radioactive waste and such spent nuclear fuel as may be disposed of in order to protect the public health and safety and the environment" and further that "high-level radioactive waste and spent nuclear fuel have become major subjects of public concern, and appropriate precautions must be taken to ensure that such waste and spent fuel do not adversely affect the public health and safety and the environment for this or future generations" (Nuclear Waste Policy Act of 1982). The Nuclear Waste Policy Amendments Act of 1987 established Yucca Mountain as the site for a deep geological repository to be prepared in the United States.

The Department of Energy's (2003) Office of Civilian Radioactive Waste Managment describes the natural barriers provided by the Yucca Mountain Project:

"* The surface soils and the natural physical shape and configuration of the mountain and its geologic environs (i.e., topography) — which limit the ability of water to infiltrate the surface

* Unsaturated rock layers above the repository level — which limit the ability of water to move down into the repository's emplacement tunnels
* Unsaturated rock layers below the repository level — which limit transport of radionuclides that might escape from repository tunnels
* Volcanic rocks and water-deposited clay, silt, and sands (alluvial deposits) below the water table — which limit radionuclide transport in the saturated zone"

The same factsheet lists the major engineered barriers as follows:

"* Drip shields — which limit the ability of water to contact the waste package
* Waste packages — which limit the water contacting the actual waste forms inside
* Cladding (corrosion-resistant metal tubes that contain the ceramic fuel pellets) — which limits the water contacting the commercial spent nuclear fuel portion of the waste
* Solid waste forms — which limit the rate of radionuclides picked up by any water that does contact the waste
* Inverts (the floors of stainless steel and crushed volcanic rock added to the emplacement tunnels) — which limit the rate of release of radionuclides to the natural barriers."

The radioactivity of the source term will decrease exponentially over the millenia, but certain long lived radionuclides will remain present in significant amounts far into the

future. The YMP was originally given a 10,000 year compliance requirement to keep doses under 15 mrem per year to a reasonably maximally exposed individual. In 2004 that standard was challenged and the Court of Appeals for the District of Columbia Circuit found that 10,000 years was not "based upon and consistent with the recommendations of the National Academy of Sciences" and required the EPA to consider longer timescales (Nuclear Energy Institute v. EPA 2004). The basis of the court's decision was the National Academy of Science (1995) statement that the time of compliance with the standard should be the time of peak dose "within the limits imposed by the long-term stability of the geologic environment, which is on the order of one million years." In 2008 the EPA published an Amendment of the Radiation Protection Standards for Yucca Mountain (Federal Register 2008) which established a two tiered compliance system; for the first 10,000 years doses must be kept under 15 mrem y^{-1}, from 10,000 years to 1,000,000 years doses must be kept under 100 mrem y^{-1}. The metric used to meet the standard was also changed, from the median predicted dose to the mean predicted dose, which as is shown in figure 1-1 will be a more restrictive standard to meet.

Over timescales of hundreds of thousands of years engineered components of any repository will degrade and waste will be exposed to groundwater and through the groundwater to the wider environment; these are timescales in which geological processes like volcanism and climate change become significant. Predicting the ways in which a system is expected to fail is essential to predicting the doses to which those

failures will lead. Figure 1-1 shows the predicted dose to nearby reasonably maximally exposed individuals (RMEI) from the YMP up to one million years. The RMEI is what it sounds like; these predicted doses are not intended to be valid for the average member of a far future community near the site of the ancient YMP. Instead, the goal is to predict the doses to those most affected by future contamination, who live and raise crops and livestock in the most contaminated areas, and to make decisions based on protecting these vulnerable individuals.

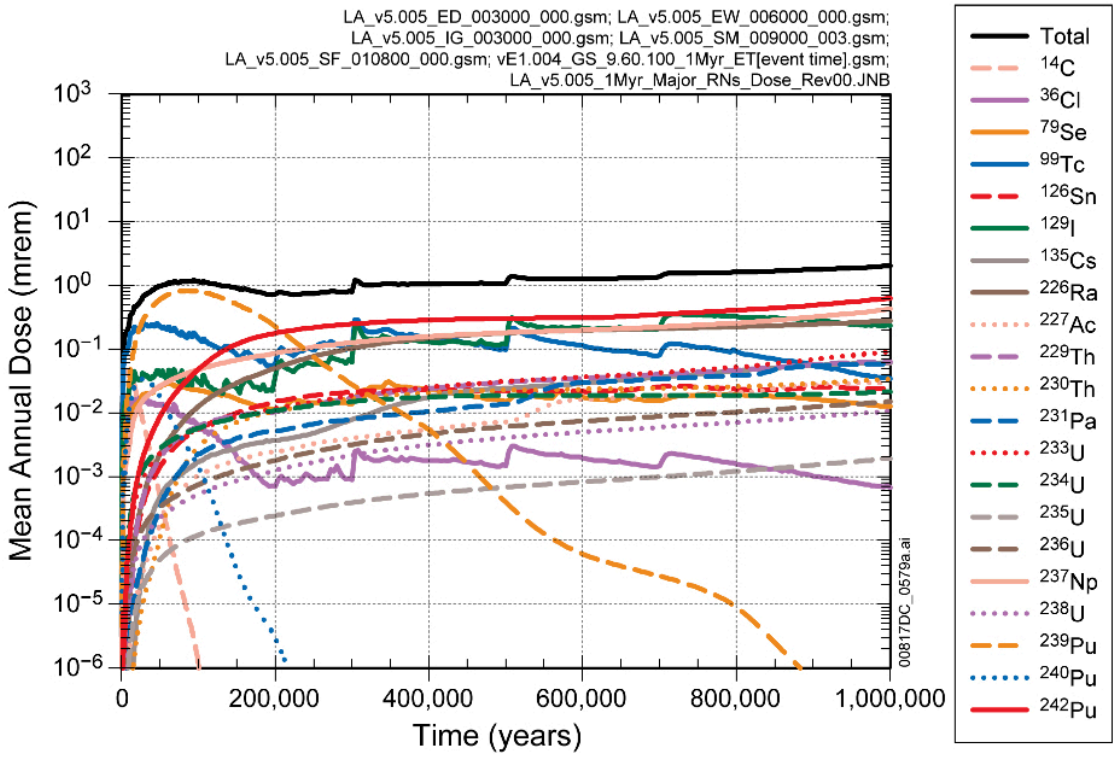

Figure 1-1 Predicted Mean Annual Dose from OCRWM's Total System Performance Assessment broken down by radionuclide (Swift 2008).

The doses in figure 1-1 are the results of a significant body of technical work, the formal process of assessing the safety of a repository is known as performance

assessment. This involves an extremely sophisticated modeling effort. Miller et al. (2000) describe the process as having the following steps:

> "1) Construction of a conceptual model which describes the system and includes all of the important processes and their couplings;
> 2) Translation of the conceptual model into a mathematical model and coding in the form of a computer program;
> 3) Verification of the numerical 'correctness' of the code;
> 4) Validation of the codes 'applicability' to the repository system to assess its predictive capabilities."

Miller et al. (2000) break down a performance assessment into a sequential chain of models, models of: canister corrosion, waste dissolution, near-field diffusive transport, far-field transport, release to the biosphere, and uptake by humans. The Electric Power Research Institute has developed a Total System Performance Assessment in support of the YMP that relies on a slightly different chain of models: Infiltration/Percolation, Seepage, Near-field containment, Near field:Source term release, Transport in the Unsaturated Zone, Biosphere, and finally dose to the reasonably maximally exposed individual (EPRI 2009). Each of these submodels contains uncertainties and is limited by incomplete understandings of the physical processes involved and these uncertainties are only magnified by the difficulty in making predictions over the timescales involved. Long and Ewing (2004) describe the problem as follows:

> "Geoscientists in this project are challenged to make unprecedented predictions about coupled thermal, hydrologic, mechanical, and geochemical processes governing future behavior of the repository, and to conduct research in a regulatory and legal environment that requires a quantitative analysis of repository performance."

As with any model, the results of a performance assessment are only as good as the parameters and assumptions that go into it. The output of a performance assessment is dependent on a staggering number of parameter values related to climate, geology, and environmental behavior, many of which are element specific, even crop specific in the biosphere models. These parameters are usually drawn from various sources in the literature and are known to different levels of certitude; for some elements and some parameters, good data does not exist. This paucity of data was demonstrated in a paper presented to Waste Management 2011 that grew out of this dissertation work whose abstract stated in part:

> "The parameters chosen for one recent performance assessment are investigated and the sources of the original data are examined. Of the 538 parameters followed, 139 (26%) reference at least one peer-reviewed article, 210 (39%) reference an institutional publication, 140 (26%) have no reference, and 49 (9%) are justified or derived internally by the case study's authors." (Higley et al. 2011)

The paper went on to note that the pedigree of the values used is often relegated to appendices when it is included at all. In the case study a number of values in recent

iterations of a performance assessment appeared to have come from the literature; however after following several nested layers of citations to an earlier document where the numbers first appeared, there is no source or justification provided for the value. This is obviously not an ideal situation: when questions of human health and safety are being considered, areas where data are lacking and further investigation is merited should be highlighted as requiring further investigation and not obscured. Presenting and defending the decisions made make a stronger safety case than masking them.

Acknowledging the difficulties in performance assessment can be challenging. The key in building a defensible safety case is transparency; it should be clearly stated both where model parameters came from, and why those parameter values are appropriate to the case at hand. In cases where there is a lack of data, there are three strategies that should be followed: using data from a chemical analogue suspected (or better, demonstrated) to show similar behavior, commisioning better data, and making conservative assumptions. Conservatism can make some allowances for uncertainty and insufficient data through tactics like choosing the most conservative value when a range of possible values exists in the literature or neglecting entirely processes that are poorly characterized and would act only to increase repository safety (Miller et al. 2000, Smith and Kato 2009). This use of conservatism should be tempered by knowledge; too much conservatism could distort the relative importance of different pathways, leading to improper allocation of resources and poor decision-making.

The future of the YMP is in doubt. Ewing and von Hippel (2009) describe the current state of affairs in a recent editorial in *Science*: "The recent action to shelve Yucca Moutain as the potential geologic repository for U.S. 'spent' nuclear fuel and high-level nuclear waste brings to a close a 30-year effort to develop and implement a policy for nuclear wastes in the United States." The apparent demise of the YMP does not obviate the need for a repository and the federal government is still obligated to establish one. The Disposal Subcommittee of the Blue Ribbon Commission on America's Nuclear Future has concluded that geologic disposal is still the best method of dealing with nuclear waste: "The United States should proceed expeditiously to develop one or more permanent deep geological facilities for the safe disposal of high-level nuclear waste…geologic disposal in a mined repository is the most promising and technically accepted option available for safely isolating high-level nuclear wastes for very long periods of time" (BRC 2011). Wherever a repository is eventually established, it will face many of the same technical hurdles as the YMP. This proposal will continue to refer to work produced in support of the YMP since those documents represent the best practices for repository development in the United States, with the understanding that the work described here would likely be as relevant to a new repository as to the YMP.

Performance assessments around the world have recognized that there are gaps in the data that is needed to populate performance assessment models. This proposal seeks to eliminate some of those gaps by providing element and crop specific data for the foliar

interception and translocation and radioactive chlorine. The dissertation is divided into five sections and an appendix: section two reviews the relevant literature, section three establishes the research objectives methods and approach, section four presents and discusses the experimental results. Section five contains the conclusions of this project. The appendix contains the plant by plant results for all experimental groups.

2 Background

2.1 Chlorine

Chlorine is the element with the symbol Cl and atomic number 17. First isolated in 1774 by C. W. Scheele it was named by Humphrey Davy (1811) who determined Scheele's dephlogisticated muriatic acid air was a new element. The word chlorine is from the Greek λωρός or khlôros meaning pale green after the color of diatomic chlorine gas. There are two stable isotopes of chlorine, ^{35}Cl and ^{37}Cl, which have a relative abundance of 75.77% and 24.23% respectively. Chemically chlorine is in group 17, the halogens, or fluoride family. Halogen comes from the latin halo (salt) and gen (creating) because the halogens readily form salts. Chlorine gas is too volatile to persist in nature and in the geosphere its only stable oxidation state is the anion chloride; Cl^-. As chloride it is abundant in the environment, making up 1.3% of the mass of seawater and constituting an essential component in living systems. The conventional wisdom is that chloride's behavior in soil is conservative; soil organisms do not chemically alter it and it shows little if any sorption to soil surfaces. This renders it very mobile in the environment and it has frequently been used as a groundwater tracer (Brady and Weill 1996, Long and Ewing 2004, White and Broadley 2001). Ogard (1988) provides some evidence that chloride may move slightly faster than tritiated water due to anion exclusion. This anion exclusion is attributed to the generally negative charge of soil structures making certain small pore spaces inaccessible to the negatively charged chloride. Öberg (1998) has questioned

assumptions of completely conservative chloride behavior and produced some data arguing for the formation of organic chlorine in soil. The standard assumption made in the United States and most of Europe for ^{36}Cl in a waste context is that waste related chlorine will be found in a chloride form and will remain chemically unaltered (Limer et al. 2009). An essential nutrient for plant life, chloride deficiency is seldom observed because chloride is ubiquitous in soils worldwide, though levels of chloride in soil tend to decline with distance from the sea (Coughtrey et al. 1983).

2.2 ^{36}Cl

Chlorine-36 is one of the significant radioactive isotopes of chlorine: it is a beta emitting radionuclide with a half-life of 301,000 years. This isotope beta decays 98.1% of the time to ^{36}Ar with a maximum beta energy of 0.709 MeV, 1.9% of the time it undergoes electron capture to ^{36}S (JAERI 2009). There are cosmic, terrestrial, and anthropogenic sources of ^{36}Cl in the environment. The three natural means of production are: "cosmic ray spallation of ^{40}Ar in the atmosphere, interactions between cosmic radiation and Cl, Ca, and K in near-surface rocks and soils, and activation of stable ^{35}Cl in the subsurface by naturally produced thermal neutrons." About two thirds of this activity is due to production in the stratosphere (Broadley and White 2001).

Cosmogenic nuclides, including ^{36}Cl, are often used in geological dating and for studying geomorphic processes. Its conservative behavior in the environment and long half-life make ^{36}Cl in its chloride form particularly suitable for these analyses. Dating

on geological timescales requires knowledge of natural rates of ^{36}Cl production and a variety of techniques address this issue. Plummer et al. (1997) have published data from fossilized urine in ancient pack rat middens indicating atmospheric production of ^{36}Cl was a factor of two greater 11,000 years ago. Anthropogenic sources can also be used for studying geological processes. The most attention ^{36}Cl has received in the United States has been a result of anthropogenically produced "bomb pulse" chlorine resulting from atmospheric testing in the 1950s and 1960s percolating through the terrain near Yucca Mountain faster than expected. The United States Geological Survey at the request of the Department of Energy worked with Lawrence Livermore National Lab and Atomic Energy of Canada Limited to produce a *Chlorine 36 Validation Study at Yucca Mountain Nevada* addressing the issue and its implications for the YMP.

Anthropogenic sources of ^{36}Cl are mostly due to activation of stable ^{35}Cl whether the stable ^{35}Cl was found in seawater as was sometimes the case during weapons testing or whether the stable chlorine was present in material exposed to an operating reactor. Chlorine-36 was not initially considered in assessments of spent nuclear fuel because it is an activation product of stable ^{35}Cl that was present only as an impurity in precursor materials. Chloride levels in light water reactor fuel have been below detection levels of 5 or 10 mg kg^{-1} and were suspected to be much lower as fuel pellets are prepared by sintering at 1600 ºC under conditions where chlorides are volatile (Sheppard et al.

1996). It was only in the 1990's that this was shown not to be entirely true, and the potential risks of ^{36}Cl received serious investigation.

When they were investigated the potential risks proved to be significant. ^{36}Cl's long half-life and environmental mobility together make it likely to still be around when there is a failure in a waste package and will be environmentally mobile once it is released. Estimates by Nirex, a waste management organization now part of the UK's Nuclear Decommissioning Authority, indicated ^{36}Cl would be the dominant nuclide in repository post-closure assessments (Nirex 1998). This led to a series of reports from Nirex investigating the ^{36}Cl contents of various types of high and low level waste using both neutron activation analysis and chemical techniques to quantify chlorine concentrations in precursor material. The conclusions of the British program were that a 275,000 m^3 reference volume of waste has between 9 and 15 TBq of ^{36}Cl; 51% of that inventory was from graphite, 30% from the fuel, 8% from the cladding, 11% from other sources (Brown et al. 1999). Shepard et al. (1996) conclude ^{36}Cl is second only to ^{129}I as the most critical radionuclide emerging from the long-term disposal of Canadian spent nuclear fuel. Scientists from Andra, the French radioactive waste management agency, predict 77% of the peak dose at the time of peak dose from their Centre de l'Aube site and 90-95% of the peak dose from a proposed graphite waste disposal facility will be from ^{36}Cl (Leclerc 2006). ^{36}Cl is not among the dominant nuclides in performance assessments of the YMP (EPRI 2009).

2.3 Chloride in Plants

Chlorine is an essential micronutrient in plants, as it is in people, although it is abundant enough that chlorine deficiency is seldom observed either in nature or agriculture. Writing in a review in the Annals of Botany, White and Broadley (2001) report that there are two reasons for a current interest in chlorine uptake and concentration by plants. The first is the study of salt tolerance. The problem of too much chlorine is more commonly encountered than the reverse and the challenge of growing crops in saline soil is a critical issue facing world agriculture. The second is ^{36}Cl's potential to be an environmental hazard emerging from radioactive waste repositories. These two areas of study overlap, but often research directed at coping with high soil salinity is not directly relevant to issues related to ^{36}Cl associated with radioactive waste.

2.3.1 Stable Chloride in Crops

The US Department of Agriculture's Salinity Research Lab's excellent *Frequently Asked Questions About Salinity* has this to say about the motivation for their work: "Irrigation inevitably leads to the salinization of soils and waters. In the United States yield reductions due to salinity occur on an estimated 30% of all irrigated land. World wide, crop production is limited by the effects of salinity on about 50% of the irrigated land area" (Department of Agriculture 2009).

Given the scope of its impact on agriculture it is not surprising the literature on salt and crops is vast. This section will by necessity discuss a fragment of that literature while developing a few ideas relevant to this project in more detail. The detrimental effects of too much salt on crops have been known since ancient times. An inscription c. 1300 BC has been found reading: "Adad-nirari, king of the universe, strong king, king of Assyria, son of Arik-den-ili, king of Assyria…conquered, burnt, (and) destroyed the city [Taidu] and sowed salt over it" (Kuhrt 1995). Although the story of the Romans salting the city of Carthage was "well known to most students" through most of the 20th century, research has revealed the story to be a modern invention (Ridley 1986). In the God's Word Translation of the Bible in Deuteronomy 29:23 divine judgment is symbolized by increased soil salinity: "They will see all the soil poisoned with sulfur and salt. Nothing will be planted. Nothing will be growing. There will be no plants in sight." "Salt ground is in Hebrew the equivalent of desert" (Ridley 1986).

High levels of salinity affect almost every aspect of plant physiology and biochemistry, significantly reducing crop yields. The primary cause is a reduction in a plant's ability to uptake water due to osmotic pressures produced by too much salt. For this reason, the effects of salt stress are identical to water stress and reduce crop yields (Munns 2002). Plants have developed a variety of methods for coping with high salinity where it has been necessary and salt tolerance varies widely among species and even cultivar. Chloride toxicity begins around 4-7 mg g^{-1} dry weight for salt sensitive species and varies from 15-50 mg g^{-1} dry weight in more salt tolerant species (White and Broadley

2001). Strategies for salt tolerance include: “limiting ion uptake at the level of the roots or by compartmentalizing ions in areas of the plant, even in cells that are away from important metabolic sites and actively growing tissues. In some cases, it appears that salts are sequestered in older leaves that are eventually shed (abscised). Some halophytes have specialized leaf cells called salt glands that excrete salt” (Department of Agriculture 2009).

Helpful in understanding the behavior of chloride in plants is a brief consideration of how chloride is transported inside the plant. This behavior can be considered in two regimes: the smaller scale of the molecular transport across plant membranes and the larger scale study of chloride’s transport through the plant as a whole. Chloride’s behavior at membrane boundaries is driven by the voltage and concentration gradients across a membrane; transport that moves in the direction of its electrochemical gradient is referred to as passive transport (White and Broadley 2001). Passive transport can be facilitated by either ‘channel’ or ‘carrier’ mechanisms. Channels are transmembrane proteins with a central hydrophilic channel, carriers are taken to be mobile proteins than can cross membranes carrying some other substance with them. Channel mediated transport is characterized by high rates and lower specificity while carrier mediated transport is known for lower rates, higher specificity, and an ability to provide active transport (Nissen 1991). Transport is considered active when the movement is opposed to the electrochemical gradient and the plant must use energy to move chloride across the membrane.

At a larger scale, movement of chloride inside the plant is effected by transport within the xylem and phloem. Xylem is a hard walled tubular tissue in vascular plants that is responsible for the movement of water and minerals from roots to shoots and other portions of the plant. Water movement through the xylem is driven by a combination of transpirational pull due to evaporation from the leaves and positive root pressure push due to osmotic pressures. Plants have small pores on their leaf and stem surfaces called stoma that allow the gas exchange necessary for photosynthesis. When these pores open during the day to facilitate photosynthesis, water can transpire out, creating tension that pulls water and any dissolved nutrients it may contain towards the photosynthizing tissue. If a plant's roots have a water potential less than that of the surrounding soil then there are positive osmotic pressures pushing water up the xylem (Öpik and Rolfe 2005).

While functioning xylem is composed of dead tissue, phloem is living tissue. Phloem serves to move a sap consisting of water and the organic nutrients and sugars produced by photosynthesis around the plant. In general the xylem delivers the raw materials and the phloem the finished goods, this is the case with chloride as well; "during the growth of the plant, Cl^- is translocated from the root to the shoot via the xylem and is redistributed between tissues via the phloem" (Broadley and White 2001).

One early paper on foliar absorption of salt by Eaton and Harding (1958) made two conclusions that are relevant here: "When the sprinkling was intermittent, the developing leaves absorbed Na^+ and Cl^- heavily…The foregoing experiments establish beyond reasonable question that the concentration of ions in the water supply were too low to cause an appreciable uptake of ions; only when the concentrations were increased in the films by evaporation were there extensive absorptions" and "all leaves showed greater accumulation of Na^+ and of Cl^- during the day than during the night." Presumably the day-night difference is due to the previously introduced stoma being open to promote photosynthesis. Similar results were reported by Sargent and Blackman (1970) who made these observations after experiments on *Phaseolus vulgaris* leaves: "In the dark the rate of entry of chloride ions up to 24 hours is constant, but in the light entry is at first slow and then more rapid. This acceleration does not occur at low temperature."

Reisenauer and Colwell (1951) concluded that concentrations of chlorine in plants are almost linearly related to chlorine soil contents, up to the level of toxicity. This is a useful property for radioecological assessments as it implies both the concentration ratio and specific activity models that will be discussed in section 2.4.1 can accurately model chloride uptake up to the level of salt toxicity.

2.3.2 ^{36}Cl in Crops

There are only scattered references to ^{36}Cl uptake in crops in the literature before the 1980s. Menzel (1965) included chlorine in the "strongly concentrated group of elements showing concentration ratios between 10 and 1000" and Bukovac and Wittwer (1957) did some early work on foliar absorption, but no large-scale investigations existed until a Nirex project at Imperial College London. Over a period of 14 years, from 1990 to 2004, a series of lysimeter and soil column experiments studied the upward migration of radionuclides in soil. These experiments included work on 8 radionuclides: ^{137}Cs, ^{134}Cs, ^{75}Se, ^{36}Cl, ^{99m}Tc, ^{125}I, ^{22}Na, and ^{60}Co. Generally their results mirror the conventional wisdom: "Overall, the clear tendency for ^{36}Cl to behave conservatively and follow the flux of moisture through the columns suggests that it is likely to be of significant importance in relation to the risk associated with its potential migration from a radioactive waste repository to the biosphere" (Wheater et al. 2007). This work resulted in a number of publications, but the recently published monograph cited here presents the data in the greatest detail.

Chlorine-36 has also been among a group of the nuclides investigated in a recent series of papers coming out of Ukraine. These papers report the results of experiments in the Chernobyl exclusion zone and focus on reporting concentration ratios for crops, though they also investigated factors such as the percentage of contamination lost during food preparation (Kashparov et al. 2005, Kashparov et al. 2007a, Kashparov et al. 2007b).

Kashparov et al. (2005) report more that half of ^{36}Cl activity in the arable layer of soil passed into radishes, lettuce, and the aboveground parts of wheat during a single growing season. Colle et al. (2005) reporting additional results from the same group report: "60% of the contamination was extracted from the soils by the plants after one vegetative period."

Both of these recent series of experiments have focused on root uptake and are motivated by the extent to which ^{36}Cl is expected to dominate future doses from repositories and yet there has not been a systematic investigation of the potential for irrigation contaminated with ^{36}Cl to be intercepted and taken up into crops before it reaches the roots.

2.3.3 Foliar Uptake of Cl-36

In their comprehensive *Radionuclide Distribution and Transport in Terrestrial and Aquatic Ecosystems: A Critical Review of Data*, Coughtrey, Jackson, and Thorne (1983) state: "Soluble chlorine-36 can be absorbed by plant foliage extremely rapidly, especially when supply of chloride to the roots is restricted. Nevertheless, quantitative data concerning the extent and rate of absorption of soluble or gaseous chlorine compounds by plant foliage are practically non-existent."

No literature could be found addressing the interception of ^{36}Cl bearing irrigation water by crops. The earliest reference to the foliar absorption of ^{36}Cl in plants occurs in

"Absorption and Mobility of Foliar Applied Nutrients" by Bukovac and Wittwer (1957). The authors used different isotopes to evaluate the foliar absorption of 14 different elements. The leaves were either painted with small circular patches of radionuclides or soaked in radioactive solutions, so the experiments were not designed to consider interception. For analysis the portion of the leaf to which the isotope had been applied was excised and absorption was considered to be the activity of each isotope recovered from the portion of the plant to which the nuclides had not been directly applied. The authors note that this method neglects any activity that had been absorbed into the portion of the plant to which they had been applied at the time of sampling.

As discussed in section 2.1, many studies of foliar uptake addressing salt tolerance exist whose results may not be directly applicable to the problem of ^{36}Cl in the biosphere. Stolzy et al. (1966) report that: "Foliar absorption of salts from irrigation waters during sprinkler applications can result in accumulation of sufficient sodium and chloride ions to cause leaf burn and defoliation." They further note that intermittent irrigation is more potentially harmful as it allows salts to be concentrated through evaporation where they would otherwise be washed off with continuous irrigation.

There are reasons to suspect foliar uptake will not dominate uptake for chlorine as it does for certain transuranics; this is because it is both less likely to attach to plant

surfaces and more likely to be taken up in a competing pathway by the roots. Bergström and Barkefors (2004) writing in "Irrigation in Dose Assessment Models" for the Swedish Nuclear Fuel and Waste Management Company conclude: "initial retention on plant surfaces is very important for elements with low bioavailability and high adsorption to soil. As can be seen…surface contamination gives dominant contribution to the concentration except for Cl-36, Tc-99, I-129 and Cs-135. These radionuclides have a high bioavailability leading to high root uptake factors. Chlorine is also an anion, leading to low retention on vegetation surfaces." This quotation also emphasizes the fact that in the context of radioactive waste, the only form of chlorine being considered is the anion chloride.

Despite the recent interest in ^{36}Cl there are still significant gaps in the literature. There is no element specific data for foliar interception of chloride in the literature and only scattered data for foliar absorption. As is discussed above, the Total System Performance Assessment for the YMP suggests using iodine data for chlorine, presumably justifying this on the basis of their shared halogen status (EPRI 2006). Other modeling efforts have made similar substitutions (IAEA 1996). In addition to the research proposed here, an ANDRA sponsored project dedicated to foliar interception that will consider ^{36}Cl among other nuclides is being planned (Madoz-Escande et al. 2009).

2.4 Modeling the Biosphere: Chloride in Crops

Before delving into the details of how current models predict ^{36}Cl concentration in crops a brief review of the history of this type of uptake problem will help frame the discussion. Prior to the Chernobyl accident foodchain models usually did not take into account radionuclide transfer to humans via foodstuffs derived from semi-natural ecosystems (IAEA 1996). Although Oak Ridge National Laboratory report 6542 (1989) makes no mention of Chernobyl in its title, "Pasture Grass Interception and Retention of ^{131}I, ^{7}Be, and Insoluble Microspheres Deposited in Rain," the project's inception in 1987 can hardly be coincidence. The PATHWAY model of Whicker and Kirchner (1987) that considers foliar interception also dates from this year.

Much of the basic radioecological work done after Chernobyl has been applicable to waste management as well and has been used to inform analyses of waste repositories. The phases of a performance assessment for radioactive waste repositories as listed in the introduction were: canister corrosion, waste dissolution, near-field diffusive transport, far-field transport, release to the biosphere, and uptake by humans. This work is relevant only to the modeling of ^{36}Cl released to the biosphere and would rely on models earlier in the performance assessment for the amount of ^{36}Cl present in groundwater near the site of the repository. The scenario considered by most performance assessments is that of future human inhabitants living off the land in an

area near the former repository site using contaminated ground water to irrigate their crops (Limer et al. 2008, EPRI 2009).

Performance assessments consider the same basic scenario, but there are differences in the way models are developed from country to country due to differences in the repository site details or cultural practices. Since in spite of their differences they address similar problems, many of the organizations responsible for radioactive waste disposal have come together in cooperative fora such as BIOPROTA: "an international collaboration forum that seeks to address key uncertainties in the assessment of radiation doses in the long term arising from release of radionuclides as a result of radioactive waste management practices" (BIOPROTA website, 2009). BIOPROTA has a ^{36}Cl working group that has published a model intercomparison report examining the sources of similarities, differences, and uncertainties in the models of ^{36}Cl behavior in the environment used by different national governments (Limer et al. 2008). The group applied 11 different ^{36}Cl models from a number of countries to two scenarios in an attempt to understand where and why models diverged. A second intercomparison report that among other things considers predictions of foliar uptake where the first considered only root uptake is in the final stages of preparation. Limer et al. (2008) deal with the subjects of section 2.4.1 in greater depth than they are covered here. The second report by Limer et al. (2009) in which this author participated has been finalized and published by ANDRA; a summary was published in the *Journal of Radiation Protection* (Bytwerk et al. 2011).

Portions of the report and some of the nomenclature from the above working group are presented here. One result, reproduced below, captures the expected contributions of root and foliar uptake to crop concentrations by three different concentration ratio models for a given scenario.

Table 2-1 The respective contributions of root and foliar uptake predicted by three different concentration ratio models (Bytwerk et al. 2011).

Crop	IMARC	ERB2A	Aquabios
Cereal (total in Bq kg^{-1} fresh)	50.0	45.4	67.8
Cereal via foliar uptake	2%	2%	0.1%
Cereal via root uptake	98%	98%	100%
Leafy green vegetables (total in Bq kg^{-1} fresh)	47.8	60.9	58.5
Leafy green vegetables via foliar uptake	13%	9%	1%
Leafy green vegetables via root uptake	87%	91%	99%
Fruit (total in Bq kg^{-1} fresh)	23.1	16.0	55.8
Fruit via foliar uptake	83%	76%	2%
Fruit via root uptake	17%	24%	98%
Root vegetables (total in Bq kg^{-1} fresh)	21.6	14.1	29.0
Root vegetables via foliar uptake	3%	7%	0%
Root vegetables via root uptake	97%	93%	100%

The impact of model assumptions can also be seen in table 2-1; IMARC and ERB2A consider ^{36}Cl directly deposited on the plant surfaces and not weathered off or absorbed at the time of consumption in addition to ^{36}Cl deposited on leaves and then absorbed and translocated within the plant, whereas Aquabios includes only the latter pathway, which explains the variation in fruit results.

2.4.1 Types of Biosphere Models

The two methods for modeling ^{36}Cl covered in sections 2.4.1.1 and 2.4.1.2 are the standard for predicting the behavior of ^{36}Cl in the biosphere. There are some alternative models in use, notably the UK's NDA model detailed in Limer et al. (2008, 2009) but conventional models based on concentration ratios and specific activity models based on isotopic dilution remain the standards.

2.4.1.1 Conventional Models

Conventional models, such as the EPRI IMARC model used for Yucca Mountain and the ERB2A BIOMASS model are based on Concentration Ratios, sometimes reported as Transfer Factors. Limer et al. (2008) list three assumptions underlying conventional models.

> "(1) A radionuclide residence time and accumulation in root-zone soil with consideration of infiltration into deeper soil by water; (2) an instantaneous equilibrium soil-water distribution coefficient to determine the mobile fraction; and (3) a fixed plant to soil concentration ratio, through which the concentration in the plant is determined as a result of root uptake."

Concentration ratios are among the most commonly used methods of predicting radionuclide levels in biota based on their levels in the environment.

$$\text{Concentration Ratio} = \frac{\text{Concentration in plants}}{\text{Concentration in media (soil, water, sediments)}} \quad (1)$$

These are empirical values from field or laboratory experiments and the values are often published in the literature without many details about the conditions under which they were obtained (Higley and Bytwerk 2007).

Numbers in the literature often have dubious pedigrees. PNNL report 13421, "A Compendium of Transfer Factors for Agricultural and Animal Products," gives a CR value of 70 for chlorine (Staven et al. 2003). The "primary reference" for their value is Baes et al. (1984), "A Review and Analysis of Parameters for Assessing Transport of Environmentally Released Radionuclides through Agriculture," which is ORNL report 5786. Examination of the ORNL report finds the source of the CR value of 70 was obtained indirectly and theoretically by assuming an average soil chloride concentration of 100 ppm and considering the range of reported concentrations in plants, which range from 2,000 to 23,000 ppm. Baes et al. chose this indirect method because the two values they could calculate based on data in the literature were deemed unsuitable. One of the sources deemed unusable, "Elemental Content of Tissues and Excreta of Lambs, Goats, and Kids Fed White Sweet Glover Growing on Fly Ash," had data implying a concentration ratio of 2.1 (Furr et al. 1978). Kashparov et al. (2007) found CR values of between 18 and 377 depending on plant and soil type.

Conventional models take interception of contaminated irrigation water into account with a term that follows the concentration factor term, the concentration in the plant is the sum of contributions from root uptake and uptake as a result of interception. Below is the soil/plant/irrigation water submodel used to calculate crop concentrations in the EPRI IMARC model and the IAEA's ERB2A BIOMASS model written using the EPRI nomenclature.

$$C_{\text{crop}} = C_{\text{root uptake}} + C_{\text{foliar uptake}} \tag{2}$$

where:

- C_{crop} is the radioactive chloride concentration in the crop, (Bq kg^{-1});
- $C_{root\ uptake}$ is the radioactive chloride concentration in the plant attributable to root uptake, (Bq kg^{-1});
- $C_{foliar\ uptake}$ is the radioactive chloride concentration attributable to intercepted activity, (Bq kg^{-1}).

$$C_{\text{root uptake}} = \frac{(F_{\text{int}} CF_{\text{crop}} + F_{\text{soil}} S_{\text{crop}}) C_{\text{soil}}}{(1-\theta_{\text{t}})\rho} \tag{3}$$

where:

- C_{soil} is the soil concentration (Bq m^{-3} (dry weight of soil));
- CF_{crop} is the concentration factor from root uptake for the crop (Bq kg^{-1} (fresh weight of crop) / Bq kg^{-1} (dry weight of soil));
- S_{crop} is the soil contamination on the crop (kg (dry weight soil) kg^{-1} (fresh weight of crop));
- θ_t is the total porosity of the cultivated soil compartment (-);
- ρ is the grain density of the cultivated soil compartment (kg m^{-3});
- F_{int} is the fraction of the internal contamination associated with the edible part of the plant at harvest that is retained after food processing has occurred (-); and
- F_{soil} is the fraction of external soil contamination on the edible part of the crop retained after food processing (-).

$$C_{\text{foliar uptake}} = F_{\text{intercept}} V_{\text{irr}} C_{\text{water}} \left(\frac{(1 - F_{\text{abs}}) e^{-\lambda_w T} F_{\text{ext}}}{Y} + \frac{F_{\text{abs}} F_{\text{int}} F_{\text{transloc}}}{Y} \right) \qquad (4)$$

where:

- $F_{intercept}$ is the fraction of irrigation water that is intercepted by the crop (-);
- V_{irr} is the volume of irrigation water applied to the cultivated soil (m^3 y^{-1} per m^2 soil);
- C_{water} is the concentration of ^{36}Cl contamination in the irrigation water (Bq m^{-3});

- F_{abs} is the fraction of intercepted radionuclide initially deposited onto the plant surfaces that is absorbed from those surfaces (-);
- λ_w is the weathering rate (y^{-1});
- T is the time between the irrigation event and harvest (y^{-1});
- F_{ext} is the fraction of external contamination from foliar interception that is retained on the edible part of the crop after food processing (-);
- $F_{transloc}$ is the fraction of absorbed activity that is translocated to the edible portion of the plant by the time of harvest (-);
- Y is the biomass yield (kg m^{-2} y^{-1} fresh weight).

2.4.1.2 Specific Activity Models

Specific activity or isotope ratio models are used for radionuclides that have stable isotopes that are abundant in the environment like ^{3}H and ^{14}C. The critical assumption made in these models is that stable and radioactive isotopes quickly reach equilibrium in all compartments. Specific activity models often lead to data problems, as tracking all sources of stable chloride into the system can be difficult. It is also more applicable to long term than short-term assessments due to seasonal variations in soil chloride contents and the time that is required to reach equilibrium (BIOPROTA 2006).

$$\frac{^{36}Cl_{plant}}{stable\_Cl_{plant}} = \frac{^{36}Cl_{plant-environment}}{stable\_Cl_{plant-environment}} \quad (4)$$

Where:

- $^{36}Cl_{\text{plant}}$ is the concentration of ^{36}Cl in the plant (Bq kg^{-1});
- $stable\_Cl_{\text{plant}}$ is the concentration of stable chlorine in the plant (mg L^{-1});
- $^{36}Cl_{\text{plant-environment}}$ is the concentration of ^{36}Cl in the plant environment where the plant environment consists of the water and soil from which the plant draws its substance (Bq kg^{-1}); and
- $stable\_Cl_{\text{plant - environment}}$ is the concentration of stable chlorine in the plant environment (mg L^{-1}).

The Andra specific activity model AquaCl36 as described in Limer et al. (2009) is included below with an excerpt explaining it to illustrate how an isotope ratio model is applied.

$$IR_{\text{soil}} = \frac{T_{\text{irr}} C_{\text{irr}}}{T_{\text{irr}} \cdot C_{\text{irr-stable}} + T_{\text{rain}} \cdot C_{\text{rain-stable}} + T_{\text{fert}} \cdot fert_{\text{stable}}} \quad (5)$$

Where:

- T_{rain} is the precipitation rate (L m^{-2} y^{-1});
- T_{fert} is the fertiliser application rate (kg m^{-2} y^{-1});
- C_{irr} is the concentration of ^{36}Cl in irrigation water (Bq L^{-1});
- $C_{\text{irr\_stable}}$ is the concentration of stable chlorine in irrigation water (mg L^{-1});

- C_{rain_stable} is the concentration of stable chlorine in rain water (mg L^{-1}); and
- $fert_{stable}$ is the concentration of stable chlorine in fertiliser (mg kg^{-1}).

"With the isotopic ratio known, percolation can be disregarded, because it has no impact on the ratio. But it has to be kept in mind that water fluxes of rain and irrigation are on a yearly basis; when percolation happens preferentially during high precipitation months (preferential loss of stable Cl) and no percolation happens during the plant growth period during which irrigation (thus contamination with Cl-36) occurs (preferential retention of Cl-36), percolation does impact the isotopic ratio. In this case, the uncertainty on the IR increases" (Limer et al. 2009).

2.4.2 Foliar Uptake

In their *Role and Uncertainty of Foliar Transfer in Radiological Impact Assessments: State of the Art and Future Actions* Madoz-Escande et al. (2009) provide an excellent motivation for the study of foliar uptake:

> "The deposition of radionuclides on vegetation and soil represents the starting point for their transfer in the terrestrial environment and in food chains. Interception is the second parameter entering the model and is defined as the fraction of a radionuclide deposited by wet deposition that is initially retained by the vegetation. Although the activity retained is subsequently removed by weathering to the soil and, the fraction that is initially intercepted is a very important quantity in all radioecological models…Translocation describes the

> systematic transport of radionuclides in the plant subsequent to foliar uptake. Translocation has no or very little influence on the long-term-fate of radioactivity in the environment, since it only describes the distribution of radionuclides within the plant subsequent to foliar deposition and absorption by the leaves. However, for estimating radionuclide concentrations in foods and for the assessment of doses to man, the systematic transport of radionuclides in a key issue."

The following two subsections develop concepts of interception and translocation in greater detail and consider how they have been modeled in the past.

2.4.2.1 Foliar Interception

A recent and comprehensive review by Pröhl (2009) *Interception of Dry and Wet Deposited Radionuclides by Vegetation* found six references for interception factors for eleven radionuclides and goes on to describe the common ways interception fractions are used in the literature; his nomenclature has been adopted here.

The simplest term is the unitless interception fraction (f) which is the ratio of the activity initially retained by the standing vegetation A_i (Bq m^{-2}) to the total activity deposited A_t (Bq m^{-2}).

$$f = \frac{A_i}{A_t} \tag{5}$$

Many sources report mass interception fractions f_B ($m^2\ kg^{-1}$):

$$f_B = \frac{f}{B} \tag{6}$$

where B is the standing plant biomass (kg m^{-2}, dry mass). Taking the standing plant biomass into account allows for the consideration of plant development, it is common sense that irrigation water is more likely to hit plant material the more plant material there is to hit and a deposition event that occurs when the plants are tiny shoots will see less interception than the same deposition on a fully mature crop.

For this same reason a metric known as the Leaf Area Index (LAI) is often used. The LAI is the ratio of the single sided leaf area (m^2) to the soil area (m^2).

$$f_{LAI} = \frac{f}{LAI} \tag{7}$$

LAI is an important parameter in many types of models; in climatology it is often a critical parameter for models of canopy response to climate change (Jonckheere et al. 2004) while from a radioecological context it represents the: “key characteristics of the rain/plant interface” better than the standing biomass (Pröhl 2009). It can be measured through direct methods like collecting leaves and measuring their area, or indirectly by

optical means such as measuring light intensities inside and outside of a canopy or through the use of hemispherical photography (Jonckheere et al. 2004).

One unfortunate result of this profusion of metrics is that often papers will report only one number and leave out crucial information needed to get other numbers. An equation that is widely used to predict the interception fraction in the literature from Chamberlain (1970):

$$f = 1 - e^{-\mu B} \tag{8}$$

where B is the aboveground biomass (kg m^{-2}) and μ is an empirical absorption coefficient (m^2 kg^{-1}). This equation was designed for dry deposition, but many investigators use it for wet deposition as well (Chamberlain 1970, IAEA 1996).

Table 2-2 Mass Interception Fraction (f_B) data from Angeletti and Levi as reported in IAEA (1996).

Amount of Rainfall (mm)	I^- (m^2 kg^{-1} dry mass)	H_2O (m^2 kg^{-1} dry mass)	Sr^{2+} (m^2 kg^{-1} dry mass)
1	4.3	6.2	7.6
2	1.6	4.3	5.1
4	1.1	1.8	4.8
8-12	0.6	1.2	4.2
16-22	0.27	0.45	1.3

Remembering that iodine data have been used in the absence of chloride data, Table 2-2 provides iodine mass interception fractions. The behavior displayed in Table 2-2 is consistent with conclusions made by Hoffman et al. (1989): "The results from this study indicate that soluble ^{131}I is transferred less readily from rain to vegetation than either a reactive cation such as $^{7}Be^{2+}$ or insoluble particles. During a continuous application of rain, ^{131}I appears to be lost from the plant with the runoff of water, regardless of the intensity of rain application. Thus, the interception and initial retention by vegetation of ^{131}I deposited in a single rain event are inversely related to the amount of rain applied."

2.4.2.2 Foliar Absorption and Translocation

As far back as 1964 it has been recognized that the "reproducibility and interpretation of results of foliar absorption studies, especially those relating to possible mechanisms of uptake, have been difficult. Problems of application, control of temperature, leaf surface moisture, relative humidity, concentration of external solution, and in distinguishing between adsorption and absorption and from transport, have been difficult to resolve" (Jyung and Wittwer 1964). Difficult enough that the IAEA's recent (2009) *Quantification of Radionuclide Transfer in Terrestrial and Freshwater Environments for Radiological Assessments* includes the phrase "In contrast with other transfer parameters, particularly with regard to soil-plant transfers, no experimental method has been standardised so far. Hence, there are as many experimental protocols as there are experiments, and results remain very heterogeneous" (IAEA 2009).

Much of the recent work in retention and translocation has come out of French researchers who have gone some way towards developing standard methods (Henner et al. 2005, Madoz-Escande 2009, Colle et al. 2009). Below are a number of metrics propounded in Henner et al. (2005) in work with bean plants. Their experimental procedure (in beans) was to soak the first two trifoliate leaves of each plant in a contaminated solution for three hours, then remove the solution and allow the plants to develop until pods had formed and were ready to harvest. Their first metric is the retention factor:

$$\text{retention factor} = \frac{\text{leaf available activity (Bq)}}{\text{activity of the whole plant at harvest (Bq)}} \tag{9}$$

The leaf available activity is a concept pioneered by Müller and Pröhl (1993) and is defined as the activity contained in a 1mm thick water layer on the surface of the leaf during soaking. This layer is meant to be representative of the thickest layer of water remaining on the surface of a leaf after rain or irrigation.

The second is the soil to pod transfer factor:

$$\text{solution to pod transfer factor} = \frac{\text{activity of plant biomass at harvest (Bq kg}^{-1})_{dw}}{\text{activity of contaminating solution (Bq l}^{-1})_{dw}} \tag{10}$$

The third is the global translocation factor:

$$\text{global translocation factor} = \frac{\text{activity of initially uncontaminated biomass (Bq kg}^{-1})_{dw}}{\text{activity of contaminated leaves (Bq kg}^{-1})_{dw}} \quad (11)$$

The fourth is the leaf to pod translocation factor:

$$\text{leaf to pod translocation factor} = \frac{\text{activity of bean pods (Bq kg}^{-1})_{dw}}{\text{activity of contaminated leaves (Bq kg}^{-1})_{dw}} \quad (12)$$

A review of the literature found scattered references to foliar uptake in the radioecological literature, but a great body of work in the literature of the plant science community. The absorption and translocation of herbicides in both weeds and crops is of great interest to companies such as Bayer and Monsanto who market both herbicides and crops resistant to certain herbicides (e.g. Monsanto's "Roundup Ready" crops). Glyphosate is the most widely used herbicide in the world, largely because of the marketing of glyphosate-resistant crops, and glyphosate relies on translocation in the plant for its efficacy (Shaner 2009). When weeds develop resistance there are powerful economic interests at stake and so there is a significant body of experimental work devoted to understanding the mechanisms and extent of foliar uptake. Some standard methodologies have been developed in the plant science community that have not appeared in the radiological literature. The general form of these experiments is to take plants, apply an herbicide radiolabelled with ^{14}C, then harvest the plants at varying

periods of time post application, usually from 2-72 hours after deposition of the radiolabelled herbicide. The plant is separated into treated and untreated sections, in some cases the untreated sections are further subdivided into further groups e.g. tissue above the treated leaf, aboveground tissue below the treated leaf, and root tissue. At harvest the treated leaf is washed and usually this wash water is retained. The leaf wash is performed with a variety of solutions, most commonly water (Brewer and Oliver 2009), or mixtures of water with additives such as polyoxyethelene sorbitan monolaurate (Everman et al. 2009) or methanol (Troxler et. al 2007) where additives were deemed necessary to remove unabsorbed herbicide.

There are established practices for experimentation and reporting. In "Leaf Wash Techniques of Estimation of Foliar Absorption of Herbicides" Devine et al. (1984) wrote:

> "Foliar absorption is an important aspect of many experiments in herbicide physiology. Determination of the amount of an applied herbicide absorbed by treated plants is commonly measured by one of two methods. One method involves washing the treated zone or removing it completely, quantifying the herbicide present in all other plant parts, and calculating absorption as the difference between total herbicide recovered and the amount applied. The second method involves washing the treated leaf and quantifying the amount of herbicide in the wash solution. Absorption is determined by expressing the

amount of herbicide in the leaf wash as a fraction of the total amount recovered from all plant parts plus the leaf wash."

The first method utilized in this work takes the activity recovered in the plant and divides it by the total activity recovered from the plant and the wash water. This is denoted the Recovered over Recovered (ROR) absorption method.

$$\text{ROR} = \frac{\text{rec. activity in plant (Bq)}}{\text{rec. activity in plant and washwater (Bq)}} \tag{13}$$

Similar to the first method is the Recovered over Applied (ROA) absorption method, except the activity ostensibly applied replaces the activity recovered in plant and wash water.

$$\text{ROA} = \frac{\text{rec. activity in plant (Bq)}}{\text{activity applied (Bq)}} \tag{14}$$

In reviewing the more recent literature a third method was discovered. The Inferred Absorption (IA) method simply subtracts the activity in the wash water from the activity deposited and calls the remainder absorbed.

$$\text{IA} = \frac{\text{(activity applied - activity in washwater) (Bq)}}{\text{activity applied (Bq)}} \tag{15}$$

Since 2000 the journal *Weed Science* has published more than 35 articles with absorption and translocation in the title. All these methods are currently in use. A cursory review of the literature found the inferring absorption method to be the most

widely adopted, found in five of ten recent absorption and translocation papers in the journal *Weed Science* (Askew and Wilcut 2002; Bukun et al. 2010; Lycan and Hart 2006; Young et al. 2003; Pester et al. 2001). Three of the remainder used the recovered over applied method (Dodds et al. 2007; Hutchinson et al. 2010; Pline et al. 2001) and two the recovered over total recovered method (Avila et al. 2007; Brewer and Oliver 2009).

The aim of most herbicide related research is focused on how the herbicide or its metabolites reaches the vulnerable sites and so most research contains absorption, translocation and metabolism work. There are two primary translocation metrics in the plant science literature, these two methods parallel the differences in equations (13) and (14). When calculating the percent translocated to a part of the plant (e.g. treated leaf, lower foliage, roots) the first method divides the activity found in the plant part by the total activity applied. The second method divides the plant part's activity by the total activity present in the plant. Both the ROR translocation method (Lycan and Hart 2006) and the ROA translocation method (Askew and Wilcut 2002) appear in the recent literature.

$$\text{ROR} = \frac{\text{rec. activity in plant part (Bq)}}{\text{rec. activity in total plant (Bq)}} \tag{16}$$

$$\text{ROA} = \frac{\text{rec. activity in plant part (Bq)}}{\text{activity applied (Bq)}} \tag{17}$$

One other translocation metric is described in the IAEA's (2009) *Quantification of Radionuclide Transfer in Terrestrial and Freshwater Environments for Radiological Assessments.* This metric is included below as the concentration ratio (CR) method that compares the activity concentration in the uncontaminated edible tissue of the plant to the activity in the foliage initially contaminated. The IAEA Tecdoc contains two variants of this metric that replace activity per unit mass with activity per unit area. In the experiments described herein plants were individually potted and the first concentration ratio method, equation 18, was deemed the most apposite.

$$\text{CR} = \frac{\text{concentration in edible plant (Bq kg}^{-1})}{\text{concentration in foliage (Bq kg}^{-1})} \tag{18}$$

$$\text{CR variant 1} = \frac{\text{concentration in edible plant (Bq kg}^{-1})}{\text{activity within 1 m}^2\text{ of foliage (Bq m}^{-2})} \tag{19}$$

$$\text{CR variant 2} = \frac{\text{activity within 1 m}^2\text{ of edible crop (Bq m}^{-2})}{\text{activity within 1 m}^2\text{ of foliage (Bq m}^{-2})} \tag{20}$$

3 Research Objectives, Methods, and Approach

3.1 Research Objectives

Given the significance of ^{36}Cl as a potential hazard and the relative scarcity of data, a number of investigators have called for further experimental work with ^{36}Cl (IUR 2006, Limer et al. 2008, Madoz-Escande et al. 2009). The 2008-2009 working group sponsored by BIOPROTA in which this investigator participated noted that no suitable foliar interception data for ^{36}Cl existed (Limer et al. 2009). There are thirty-six chlorine specific parameters that contribute to the EPRI IMARC model for the YMP but on the element specific parameter table for chloride in the EPRI report the phrase "In the absence of data for chlorine using data from iodine" appears in the reference comment field six times while "data for iodine" appears twice; no chlorine specific data related to foliar uptake is present (EPRI 2006). Even the iodine data that exist often come from references that do not report their results with a standard method. For example, as is discussed in section 2.4.2, foliar interception can be described with a unitless interception fraction or with a mass-interception fraction which takes into account the standing plant mass; one journal article might report only the mass-interception fraction, but not the standing plant mass a reader would need to convert the mass-interception fraction to the unitless interception fraction if that was the value needed by their model. This means that even when data exist they are often not available in a form that is of utility to potential users.

This project enriches the literature with experimental data for the foliar uptake of $^{36}Cl^-$ from three types of crops; cereals, root vegetables, and fruit vegetables. A further goal is to work towards community standards in experimental methods in order to produce more commensurable results. Specifically this work provides values for the following parameters under a variety of conditions in several crops:

- Interception Fraction Metrics
    - Unitless interception fraction (f)
    - Mass interception fraction (f_B)
    - Interception fraction normalized to Leaf Area Index (f_{LAI})
- Foliar Uptake
    - Absorption Metrics
        - Recovered over applied absorption method
        - Recovered over total recovered absorption method
        - Inferred absorption method
    - Translocation Factors
        - Recovered over applied translocation method
        - Recovered over total absorbed translocation method.
        - Concentration ratio translocation method

3.2 Methods and Techniques

3.2.1 Crops under Investigation

The variety of plants in the human diet and the expenses and facilities involved in conducting radioecological experiments makes it impracticable to break down the human diet by species and to determine transfer parameters for each species, even in a site specific context. Instead crops are often grouped into broader categories such as root vegetables, grains, or fruits. The argument justifying this simplification is the same as that underlying the ICRP's traditional "Reference Man" or newer "Reference Animals and Plants." It is problematic to determine the weight of any particular person's thyroid, but having an agreed upon value allows dosimetric calculations based on that assumption. Just so the intelligent use of analogue species makes it possible to proceed with risk assessment in the face of biological diversity. Three different crops from three different classes of crops are investigated in these experiments; radishes, beans, and wheat. All plants were watered and fertilized with Miracle Grow All Purpose Plant Food (14111 Scottslawn Rd. Marysville, OH 43041) as needed. Plants were maintained at 68 ˚F and received supplemental artificial lighting on a 14/10 on/off schedule.

3.2.1.1 Radishes

The radish (*Raphanus sativus*) was chosen as a representative root vegetable; they are fast growing plants and well suited to greenhouse conditions. Radishes were grown in 10'' by 10'' pots in potting soil provided by the Oregon State University Department

of Crop and Soil Science (OSU CSS). Seeds, a cultivar named Crunchy Royale, were ordered from Johnny's Selected Seeds (955 Benton Ave., Winslow, Maine 04901). Two seeds per pot were planted and thinned to one on emergence. The timeline below lays out when each group of plants was treated. Each group consisted of 6 replicates: "Interception A" is a group of 6 plants that underwent the same interception experiment on 8/12/2010, 14 days after planting.

Table 3-1 Timeline for radish experiments.

Calendar Date	Crop Age	
7/30/2010	Day 1	Planting
8/12/2010	Day 14	Interception A, Translocation A
8/17/2010	Day 19	Interception B, Translocation B, Absorption A B C
8/22/2010	Day 24	Interception C D E F, Translocation C,
8/27/2010	Day 29	Interception G, Harvest

3.2.1.2 Beans

The common bean (*Phaseolus vulgaris*) was chosen as a representative fruit. Beans were grown in 13'' by 13'' pots in potting soil provided by OSU CSS. Two seeds per pot were planted and thinned to one on emergence. In mid-December thrips were spotted on the bean plants. Ferti-Lome Triple Action Plus (Voluntary Purchasing Group P.O. Box 460, Bonham, TX 75418) applied to the plants via a mister mitigated this blight, though it remained an issue through plant harvest.

Table 3-2 Timeline for bean experiments.

Calendar Date	Crop Age	Radionuclide Applications
11/1/2010	Day 1	Planting
12/2/2010	Day 31	Translocation A, Interception A
12/13/2010	Day 42	Translocation B, Interception B
12/24/2010	Day 53	Translocation C, Absorption A B C
1/4/2011	Day 64	Translocation D
1/16/2011	Day 76	Translocation E, Interception C D E F G

Figure 3-1 Thrip damage on a bean plant and an ally against them.

3.2.1.3 Wheat

The grain chosen for these experiments was common wheat (*Triticum aestivum*). *Triticum aestivum* is the most commonly cultivated species of wheat and the variety grown was Alpowa, a variety of soft white spring wheat first released in 1994 and provided by OSU CSS. Wheat was grown in 13'' by 13'' pots in potting soil also provided by OSU CSS. Two seeds per pot were planted and thinned to one on emergence. The crop developed well, with nearly 100% germination, and grew without

hindrance until the final 30 days of development when a powdery mildew developed on the crop. Ferti-Lome Triple Action Plus applied to the plants via a mister mitigated this blight. Alpowa is known to be "moderately susceptible to mildew." (U of Idaho 2011)

Table 3-3 Timeline for wheat experiments.

Calendar Date	Crop Age	Radionuclide Applications
9/27/2010	Day 1	Planting
12/2/2010	Day 66	Translocation A, Interception A
12/13/2010	Day 77	Translocation B, Interception B
12/24/2010	Day 88	Translocation C, Absorption A B C
1/4/2011	Day 99	Translocation D
1/16/2011	Day 111	Translocation E, Interception C D E F G

Figures 3-2 and 3-3 Images of powdery mildew on wheat.

3.2.2 Experimental Facilities, Equipment, and Radionuclides

The radioactive components of these experiments were conducted in the greenhouse facility associated with the Radiation Center. Prior to applications of ^{36}Cl plants were grown in a greenhouse facility operated by OSU CSS. The analysis of the samples was

performed with a Perkin Elmer (940 Winter Street Waltham, MA 02451 USA) Tri-Carb 3180 TR/SL Liquid Scintillation Counter at the OSU Radiation Center. Some use was made of a Gas Flow Proportional Counter and ^{36}Cl standard belonging to the Radiation Center's Health Physics staff to validate the LSC results. Weights were measured with an Ohaus Explorer Pro scale; dried plants were maintained in a Fisher Isotemp 200 series oven (22 Friars Drive Hudson, New Hampshire 03051), and 2 Eppendorf Research pro Electronic Pipettes. 5-100 µl and 100-5000 µl were used. The majority of the ^{36}Cl used in these experiments was a 100 µCi ^{36}Cl solution manufactured by Amersham, now G.E. Healthcare (Pollards Wood Nightingales Lane Chalfont St. Giles HP8 4SP United Kingdom) and obtained gratis through the good auspices of Oregon State University Radiation Safety. Some experiments used ^{36}Cl from a 50 µCi stock solution purchased from G.E. Healthcare. Both solutions came as NaCl and were diluted to the appropriate concentrations with deionized water.

Figures 3-4 and 3-5 Beans and radishes growing in the Plant and Soil Science greenhouse (left) and the greenhouse associated with the Radiation Center (right).

3.2.3 Plant, Stock Solution, and Wash Water Activity Determination

The analysis procedure for all the plant samples was identical. After harvest plants were weighed, dried for at least 72 hours, and maintained at 65 ºC for later digestion. In the absorbtion and translocation experiments plants were separated into edible and non-edible plant sections. Before digestion samples were reweighed and ground to a powder. Bean and wheat samples were ground with a blender; radish samples were too small to blend and were ground by hand with a mortar and pestle (in the case of the edible radish tissue) or chopped very finely with razor blades (for radish leaves). Testing showed the water taken in from the atmosphere by dried bean tissue during the grinding and weighing process resulted in a less than 3% change in mass. It is expected that this effect was negligible in other plants as well.

Figure 3-6 Collage of the sample preparation process from harvest to drying to weighing to blending, to sample storage, to subsample for acid digestion.

Plant tissue is a challenge to analyze via LSC because cellulose is very difficult to break down. Some form of acid digestion or solubilization is necessary for activity determination. Plant sample digestions for ^{36}Cl assessment were performed using the method of Wahid et al. (1985) as cited in Thompson (1998):

1. "Prepare the solubilizing reagent by adding one volume of 70% perchloric acid to one volume of 70% nitric acid.
2. Where possible, the sample should be oven dried and then finely cut.

3. Place prepared sample (up to 200 mg) in a glass scintillation vial fitted with a poly-cone lined urea screw cap.
4. Add approximately 0.6 mL of the prepared $HClO_4$-HNO_3 reagent (1:1).
5. Digest the sample in the closed vial at 50-70 ºC for 1 hour or until an almost colorless solution is obtained.
6. Cool the vial to room temperature and add 15 mL of Hionic-Fluor.
7. Temperature and light adapt for 1 hour before counting."

After a number of trial runs 100 mg samples were chosen as providing a happy medium between increasing activity in the samples and still achieving a sufficiently complete digestion. Similarly three-hour digestion times provided a better digestion than shorter periods and were used for all the results reported here. Several digestion and solubilization methods were tested; Soluene-350, nitric acid digestion, nitric-perchloric digestion, and perchlorid acid-hydrogen peroxide digestions. The nitric-perchloric digestions were found to show the most complete digestion. This method was also significantly more efficient than a nitric only digestion used in earlier experiments where larger samples were digested under reflux as shown in figure 3-7 (Bytwerk and Higley 2009).

Figure 3-7 The original digestion setup.

Figure 3-8 A collage of the digestion process from the addition of the acid, to the heated digestion, to the resulting sample, and to analysis via LSC.

Blank digestions and spiked digestions were done to characterize the efficiency of the digestion process and the extent of quench in the final samples. Quench is a term used to refer to any loss due to the characteristics of the sample in the cocktail due, for example, to incomplete digestion of the sample, color quench due to discoloration of the digested sample, or chemical quench where the sample interferes chemically with the fluorescing of the LSC cocktail. Sample activity was determined as follows:

$$\text{sample activity (Bq)} = \frac{(CR_S - CR_B)}{\text{ChYield}} * \frac{1}{CE} \quad (21)$$

Where:

CR_S = count rate of sample;

CR_B = count rate of blank sample ;

ChYield= Chemical yield, the fraction of ^{36}Cl not lost during the digestion process;

CE = Counting efficiency, the ratio of detected ^{36}Cl (cps) to actual ^{36}Cl (Bq) present in a sample due to quench correction (color and chemical).

The chemical yield and counting efficiency terms were fused into a single correction factor. The reasoning is: assume a blank (uncontaminated) sample is spiked with 1000 disintegrations per second of ^{36}Cl before being digested. After the spiked blank has gone through the digestion process and been analyzed via LSC the background corrected result for that sample is 500 counts per second. Since it is known the actual activity present predigestion was 1000 disintegrations per second multiplying the

counts per second in the sample by two will provide the true activity, in disintegrations per second, that was present in the sample before the digestion. In terms of units, this correction factor would be disintegrations per second divided by counts per second. Because the spike occurs before the digestion this correction factor takes into account losses due to volatilization during the digestion process as well as the efficiency of the liquid scintillation counter. Four spiked digestions were done for each tissue type (edible, non-edible, undivided plant) in beans and wheat, three digestions were done for each type in radishes. The results for each plant type were averaged and used to correct the raw results reported by the liquid scintillation counter. Background counts from uncontaminated samples of the same type were subtracted where appropriate.

Chlorine-36 stock solutions and wash water from the absorption experiments was also analyzed with LSC, aliquots of the solution were taken, diluted in 18ml of Hionic-Fluor cocktail and analyzed. An example of the LSC spectrum of each type of sample is included below. The LSC is reported to be 100% efficient in detecting ^{36}Cl betas in water.

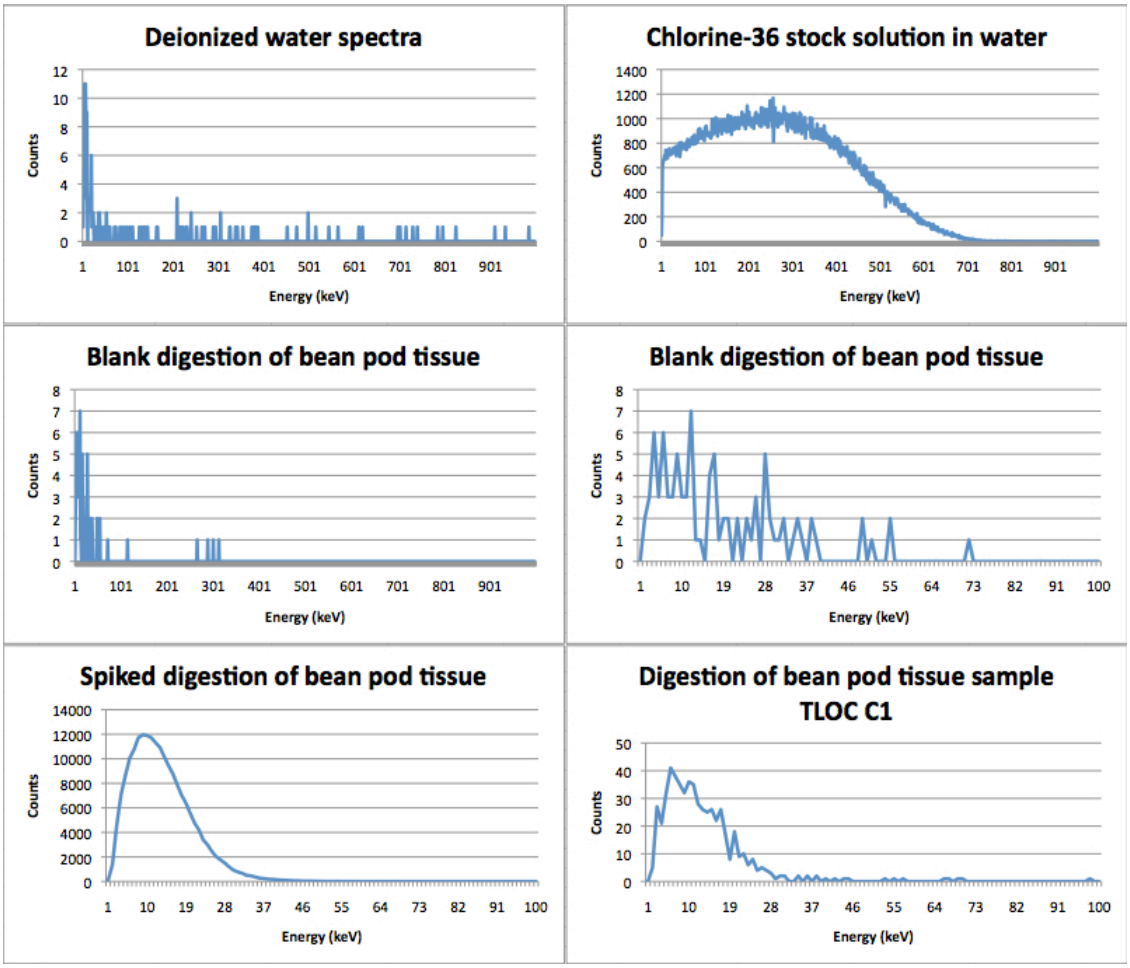

Figure 3-9 A collage of liquid scintillation spectra from each type of sample.

The first graph is of deionized water with LSC solution. The graph in the upper right is a classic beta spectrum that peaks at the maximum energy of the ^{36}Cl beta, 709keV with a maximum height at ~1/3 the maximum energy. The two middle graphs are the same graph rescaled. After digestion there is a significant amount of quench in the samples, in the blank samples background counts are of much lower energy than was seen in the deionized water samples. The final graphs are examples of ^{36}Cl beta spectra in digested samples. The curves are shifted far to the left as compared to the

unquenched ^{36}Cl in deionized water, but the total counts under the curve will not be as depressed for the relatively high ^{36}Cl beta as they would be for a beta emitter of lower energy. The left is a spiked sample with significantly greater activity that shows a smooth curve. In the bottom right is the spectrum for the bean pod tissue of the first plant in the translocation group 'C'. Its shape is the same as the spiked digestion, but due to the substantially lower count rates the curve is less smooth. Figure 3-9 is intended as a qualitative comparison, the activities present in each graph are unrelated, what is important to note is the shape of the spectra.

3.3 Foliar Interception Experimental Design

In these experiments simulated irrigation was manually applied to treatment groups of each species. Each of the three metrics for foliar interception discussed above; the unitless interception fraction, the mass interception fraction, and the interception fraction normalized to leaf area index were computed in all the experiments for each plant. Plants were transported from the Crop and Soil Science greenhouse to A126 in the Radiation Center where the interception experiments were carried out on a giant tarp in the center of the room.

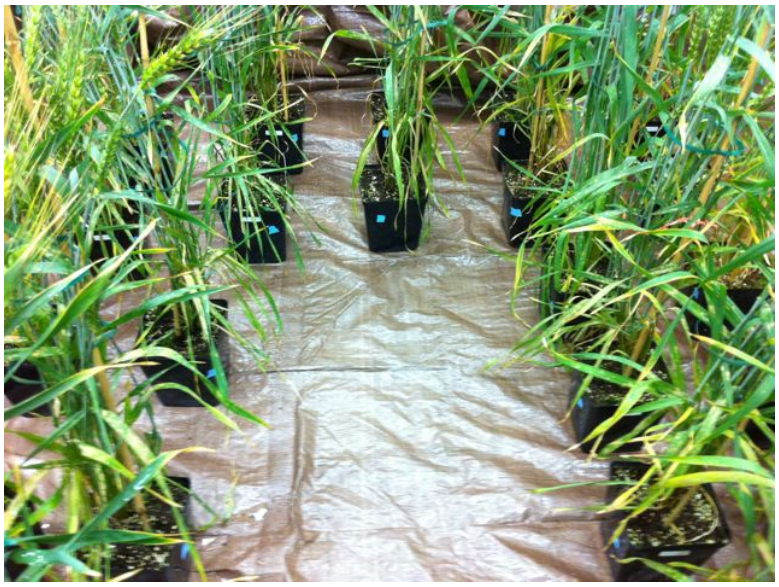

Figure 3-10 Wheat in RC A126 awaiting interception experiments.

Plants were given the appropriate deposition volume, allowed to dry for one hour or as needed, then harvested, weighed, dried, weighed again, and subjected to plant activity determination. Experiments were conducted at approximately the same time to eliminate time of day effects (e.g. during the day bean plants present as much surface area to the sky as possible but at night the leaves droop significantly).

3.3.1 Radionuclide Application in Interception Experiments

The size of the greenhouse and the presence of radionuclides in these experiments make the use of many methods of simulating irrigation impractical. It should be remembered that the scenario most commonly considered with ^{36}Cl is human irrigation from contaminated well-water so mimicking natural rainfall is not the objective. Since there are any number of methods of irrigation used in agriculture, prudence requires

conservative assumptions. In the same foliar interceptions study at Oak Ridge by Hoffman et al. (1995) cited previously is a method of deposition using a 50 ml syringe with a blunt needle to gently dispense irrigation water for the purposes of studying foliar interception and this method is adopted here. Plastic syringes of various sizes were used to gently apply the contaminated water in small droplets over the crops.

3.3.2 Leaf Area Index Determination for Interception Experiments

Measurements for the interception fraction and mass interception fraction are relatively straightforward; slightly more challenging is the LAI. The LAI in these circumstances is often measured nondestructively using specialized equipment such as the LICOR LI-3000 series area meter. These meters are expensive, costing several thousand dollars; Rico-García et al. (2009) propose a better alternative for these experiments. Their new method processes digital photography in MATLAB to calculate LAI and was found to have a correlation coefficient of above 99% when compared to standard LICOR area meter analysis.

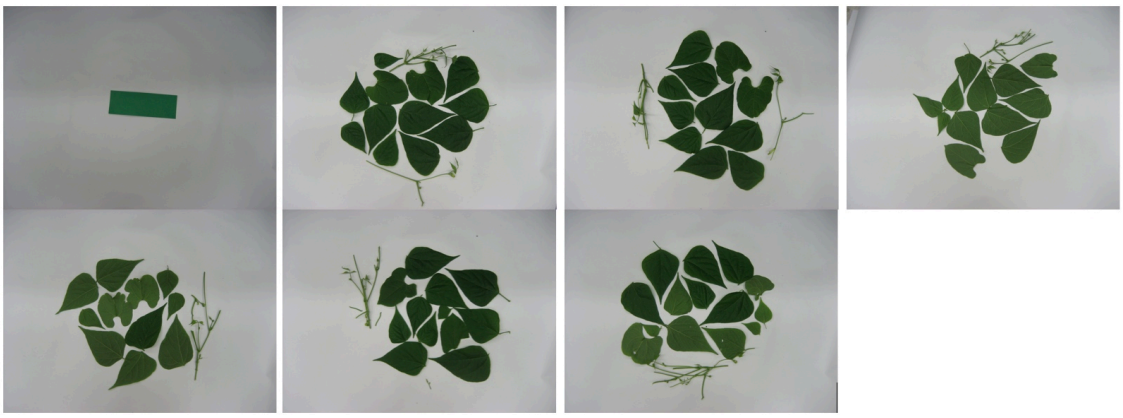

Figure 3-11 The photoset associated with the LAI index determination of bean group interception A.

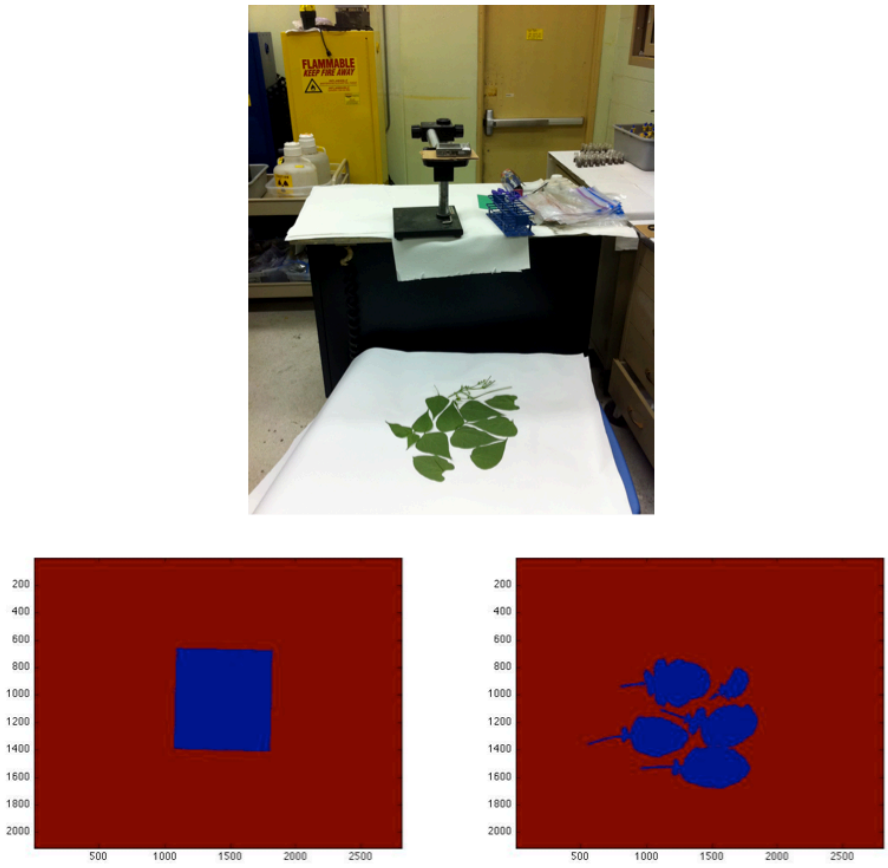

Figure 3-12 LAI index determination setup (top) and MATLAB output (bottom).

The method is to take pictures of both leaves and reference objects of known size against a white background. As long as the camera's position is fixed relative to the objects being photographed the relative area occupied in the photographs by the leaves compared to the reference object can be used to calculate the surface area of the leaves. The MATLAB script requires the manual input of calibration factors, the MATLAB .m file used is included as Figure 3-13.

```
clear;
j=0;
for i=49:55
    X=[112 104 111 116 111 i]
    Y=char(X)
    A=imread(Y,'jpg');
    C=(double(A(:,:,1))+double(A(:,:,2))+double(A(:,:,3)))/3;
    Cota_color=100;
    Pre_Area=find(C<=Cota_color);
    C(Pre_Area)=0;
    image(uint8(C))
    Area=length(Pre_Area);
    F=1.911e-004
    Real_area=Area*F;
    j=j+1;
    Varea(j,:)=Real_area;
end
Varea
```

Figure 3-13 A screenshot of the MATLAB script to compute Leaf Area Index, adapted from Rico-García et al. (2009).

There were two variables in the MATLAB script that were altered between treatment groups. The F value is the number that relates the number of pixels occupied by leaves to a surface area in square centimeters. For every experimental group a new F value was calculated based on; the previous iteration's F value, the calibration image's area calculated using the previous iteration's F value, and the true size of calibration image. The Cota_color variable was also sometimes altered to aid MATLAB in distinguishing shadow from leaf tissue and in the case of several images a photo editor, GNU Image Manipulation Program, was used to white out objects such as a plastic bag or pair of scissors that snuck into the corners of several photographs.

3.3.3 Influence of the Amount of Simulated Irrigation on Interception

These experiments are simple in character; plants at a single stage of development will be subjected to a variety of deposition volumes and the activity retained on the plant will be quantified. The common sense idea is that the greater the deposition volume,

the greater the chance that interception activity will be washed off by subsequent deposition. Increasing deposition volume is expected to depress all three of the interception metrics discussed above. Chloride is expected to behave in a manner similar to iodine and have interception fractions inversely dependent on the volume of simulated irrigation volumes as was discussed previously (Hoffman et al. 2005).

The following table lays out what deposition volume was used in each experimental group. The first number is the deposition volume in milliliters: the second number is an expression of the deposition value as a function of the pot size and can be read as a rainfall gauge would be read. So to take the first example 10ml / 1mm refers to 10 ml of water deposited on a pot 10 cm by 10 cm which would result in a layer of water in the bottom of the pot 1 millimeter deep were it empty. There were four treatment groups of radishes, five for beans and wheat.

Table 3-4 Deposition volumes for interception experiments.

	Radish	**Bean**	**Wheat**
Interception C	10ml / 1mm	17ml / 1mm	8.5ml / 0.5mm
Interception D	20ml / 2mm	34ml / 2mm	17ml / 1mm
Interception E	80ml / 8mm	68ml / 4mm	34ml / 2mm
Interception F	160ml / 16mm	136ml / 8mm	68ml / 4mm
Interception G	N/A	272ml / 16mm	135ml / 8mm

3.3.4 Influence of Developmental Stage on Interception

In these experiments the same volume of deposition will be given to plants at different stages of development. It is clear the stage of plant development will significantly impact all of the interception fraction metrics. Here the goals are both to establish metric values and to understand the merits and flaws of each. The unitless interception fraction takes no account of development unlike the other two metrics that consider standing biomass and leaf area index as indicators of plant development. Presumably as the plant grows and matures, increasing in standing biomass and leaf area index, the unitless interception fraction will also increase, though this may prove less true towards plant maturity when growth is focused on fruiting bodies and not on increasing leaf area.

3.4 Foliar Absorption and Translocation Experimental Design

These experiments trace the movement of foliar deposited ^{36}Cl into the plant, and specifically into the edible portion of the crop. In the absorption experiments the ^{36}Cl is applied and after varying periods of time the plant is harvested, the treated leaves washed to remove unabsorbed ^{36}Cl, and the activity in the edible crop and the remainder of the plant determined. In the translocation experiments the ^{36}Cl was applied in the same way only to plants of different ages and the plants will all be maintained until harvest.

3.4.1 Radionuclide Application for Absorption and Translocation Experiments

Instead of the leaf soaking method seen in the radiological literature ^{36}Cl was applied to each plant in small drops via pipette, as is the standard in the weed science community. Radishes received 0.6 ml in 75 µl drops, beans received 1.5 ml in 0.1 ml drops, and wheat received 0.6 ml in 25 µl drops. The activities deposited on each treatment group are included in the appendix. A larger deposition volume with a correspondingly higher activity was chosen for the beans, as they were significantly larger in surface area than the other crops. Depositing the activity onto the wheat was challenging. Unlike beans, which have small hairs that serve to catch water, wheat seems almost to repel it; deposited water beaded up instead of clinging to the plant. To cope with this difficulty many drops were deposited near the junction of leaves with the main stalk; these junctions served to halt and retain any escaping fluid.

Figure 3-14 Translocation experiments in wheat and radishes immediately post deposition.

3.4.2 Absorption Experiments

These experiments were very similar in design to many of the absorption and translocation papers in the journal *Weed Science*. After a deposition of ^{36}Cl plants were harvested at various intervals with the aim of quantifying the extent and speed of uptake from leaf surface to leaf interior. The treated leaves were washed with varying volumes of deionized water and both wash water and the plant, divided into edible portion and remainder, were analyzed for ^{36}Cl. DAT in the following table is days after treatment.

Table 3-5 Time after deposition before harvest for each absorption treatment group.

	Radish	**Bean**	**Wheat**
Absorption A	Harvest 1 DAT	Harvest 2 DAT	Harvest 2 DAT
Absorption B	Harvest 2 DAT	Harvest 4 DAT	Harvest 4 DAT
Absorption C	Harvest 4 DAT	Harvest 30 DAT	Harvest 30 DAT
Absorption D	Harvest 10 DAT	N/A	N/A

It was expected that ^{36}Cl would be readily absorbed through the leaves. Even in the case of herbicides, large molecules inimical to plant health, absorption percentages are often significantly over 50% (Pester et al. 2001, Everman et al. 2009).

Radish and bean leaves were washed with deionized water by gloved hand in a small plastic container; wheat plants were sealed inside a graduated cylinder and gently agitated as part of the washing process. After washing the leaves were dried and the rinsing water was retained and both were subsequently analyzed.

Figures 3-15 and 3-16 The leaf washing process in beans, wheat (left), and radishes (right).

A test of the procedure by applying the ^{36}Cl and harvesting the plant 10 seconds later found over 95% of the applied radioactivity in the wash water. This test ensures the method is functioning properly and was found in the plant science literature. Absorption factors were calculated according to equations 13, 14, and 15.

3.4.3 Translocation Experiments

In many ways this is the most interesting experiment. Plants treat many nutrients differently at different stages of development; some stages may be uniquely at risk for uptake. The experiments were started as the plants began to flower in the case of beans and wheat as it was suspected little ^{36}Cl would be translocated to the edible crop before the edible crop had begun to form. At varying intervals after the first deposition a new

group was treated with ^{36}Cl. After deposition plants were grown to maturity when they were harvested, separated into plant and edible crop, and analyzed. Translocation factors were calculated according to equations 16, 17, and 18.

4 Results

This chapter reports treatment group averages and considers the data from different perspectives. Individual plant results are available in appendix A. The results are grouped by experiment type and species and each experimental section ends with a comparison of values between species.

4.1 Foliar Interception

4.1.1 Radish Interception Results

Before considering interception proper, each interception results section will examine relationships between leaf surface area as given by MATLAB and the plant's measured aboveground biomass. Figure 4-1 evinces a strong correlation between the two in radishes.

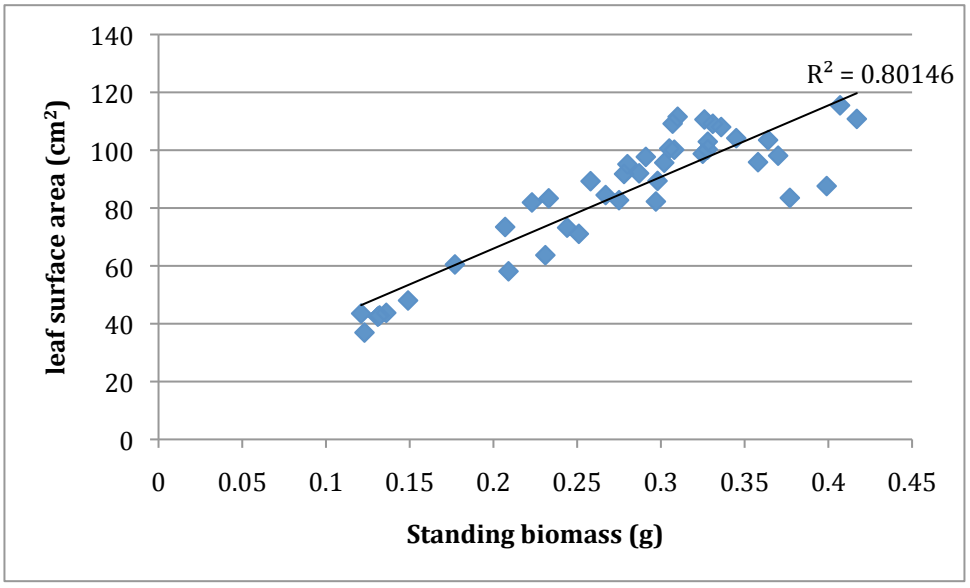

Figure 4-1 Leaf surface area vs. dry weight – all radish data.

The strong correlation is good because both f_b and f_{LAI} use these numbers to take plant development into account. Both are stand-ins for the true effect on interception of the plant's stage of growth it is fortunate that they correlate.

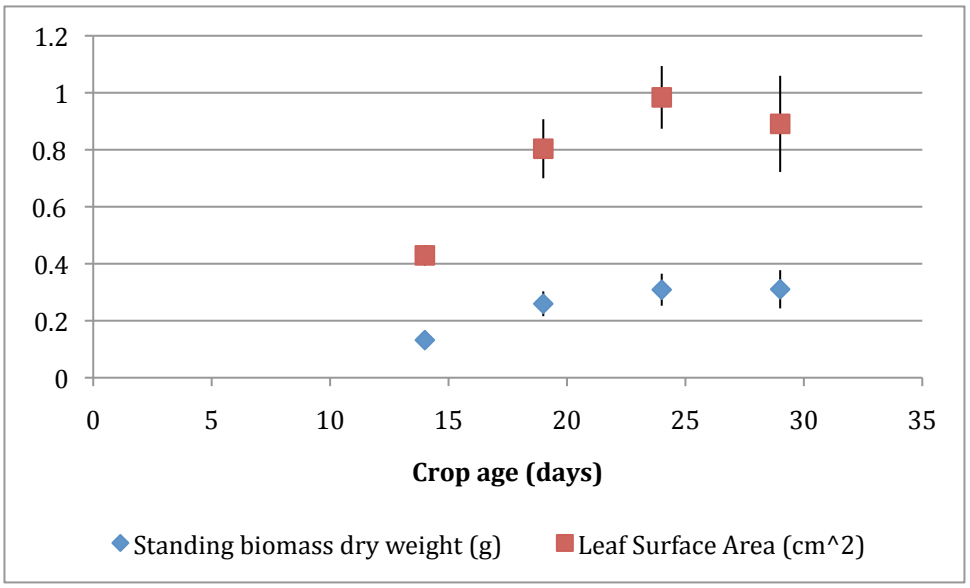

Figure 4-2 Trends in radish leaf area and standing biomass with crop development.

While they are correlated a look at the trends in each over time (Figure 4-2) shows that leaf surface area and dry weight peak over time. This is a place where the impacts of different types of plants can be seen. Radishes are root vegetables; the edible portion of the radish is underground and so does not contribute to standing biomass.

4.1.1.1 Effect of Plant Development

Table 4-1 breaks down the interception metric group averages and standard deviations for the four radish groups that received an identical volume of deposition at different stages of development.

Table 4-1 Radish interception results at varying stages of development.

Crop Age (days)		$f * 10^{-2} \pm \sigma$	$f_B \pm \sigma$	$f_{LAI} * 10^{-2} \pm \sigma$
A	14	6.0±.98	4.6±.91	14±2.2
B	19	5.8±1.6	2.2±.50	7.3±2.2
C	24	4.4±1.7	1.4±.47	4.5±1.6
G	29	5.1±.77	1.7±.34	5.9±1.2

These results are visually presented in Figure 4-3. The metrics all show the same general trend, but there are differences in the extent of the variability. The unitless interception fraction shows very little variability while f_B shows the most. Since one of the reasons for the existence of f_B and f_{LAI} is to take plant development into account and reduce variability the radish experiments do not support using these metrics over the unitless interception fraction. The decrease in unitless interception fraction with crop development is a little unusual. This is attributed to the radishes having reached a 'foliar maturity' early on, as seen in Figure 4-2 after the first group there is relatively little change in leaf surface area and standing biomass.

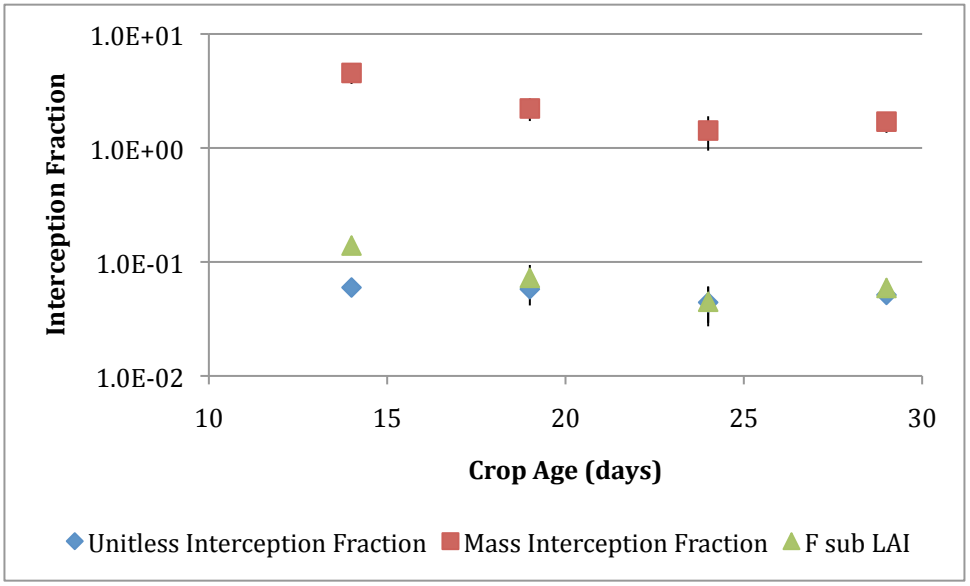

Figure 4-3 Interception fraction vs. crop age – radish groups A,B,C,G.

4.1.1.2 Effect of Deposition Volume

Table 4-2 breaks down the interception metric group averages and standard deviations for the four radish groups that received different volumes of deposition at a single stage of development.

Table 4-2 Radish interception results at with varying deposition volumes.

Dep. Vol. (ml)		$f * 10^{-2} \pm\sigma$	$f_B \pm\sigma$	$f_{LAI} * 10^{-2} \pm\sigma$
C	10	4.4±1.7	1.4±.47	4.5±1.6
D	20	3.7±1.3	1.2±.51	4±1.4
E	80	2.7±2.5	.96±.85	2.9±.2.5
F	160	2.3±1.5	.76±.51	2.4±.1.5

Here the variability between metrics is smaller, with all metrics decreasing by about a factor of 2 as the volume of deposition is increased. Figure 4-4 shows this trend and also indicates the fairly wide intragroup variation leading to high standard deviations.

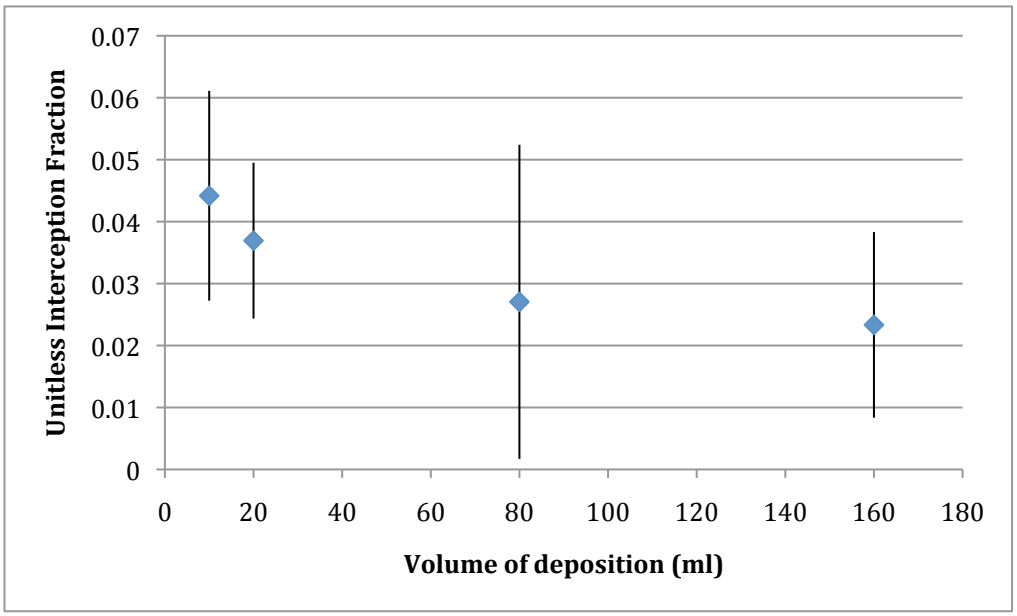

Figure 4-4 Interception fraction vs. deposition volume - radish groups C-F.

4.1.2 Bean Interception Results

Figure 4-5 shows much the same relationship as did Figure 4-1. It is again encouraging that there is a strong correlation. The data was more tightly packed to either end of the graph. One the left-hand side the behavior looks qualitatively different. The two groups, Interception A and B that make up that portion of the graph are pulled out and examined in Figure 4-6

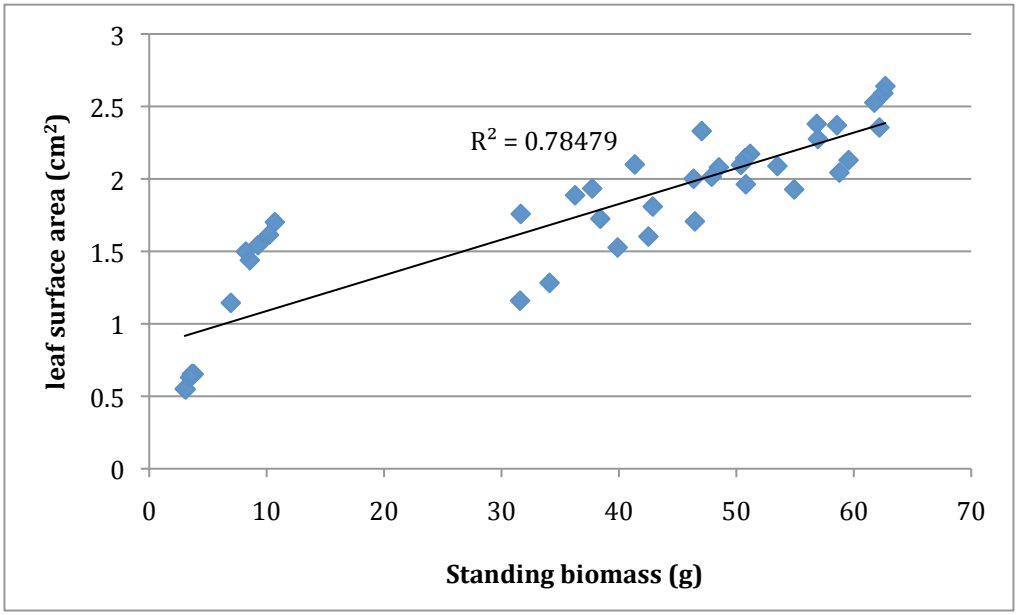

Figure 4-5 Leaf surface area vs. dry weight - all bean data

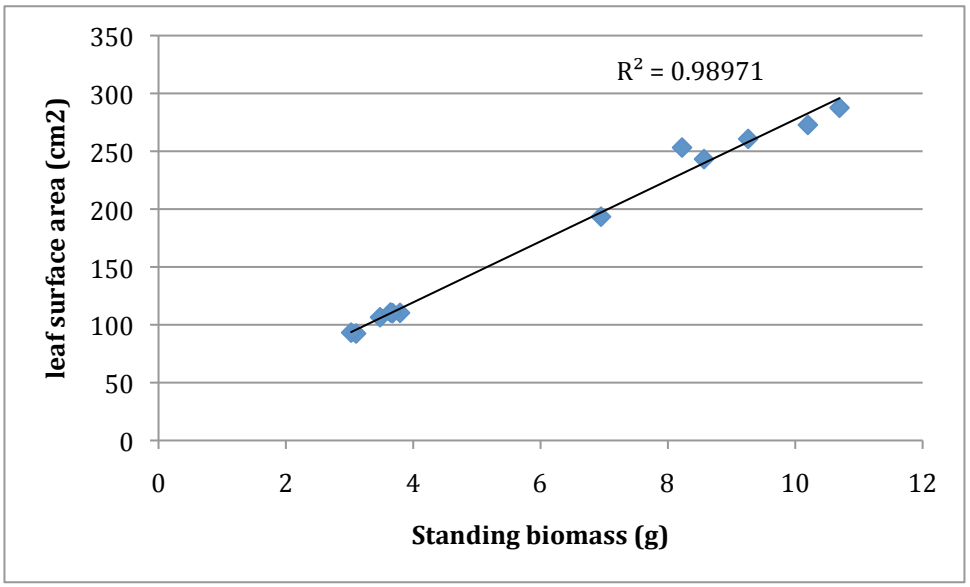

Figure 4-6 Leaf surface area vs. dry weight – bean interception groups A and B.

Interception groups A and B were the two earliest interception experiments when the plants were least developed. Figure 4-6 shows the near linearity of the standing biomass – leaf surface area relationship early in the plant's life when the majority of a

plant's energy is spent expanding and it is not simultaneously devoting resources to a fruiting body.

Something of the same relationship can be seen in Figure 4-7. Initially the leaf surface rose quickly and then more slowly, while the standing biomass reversed these trends.

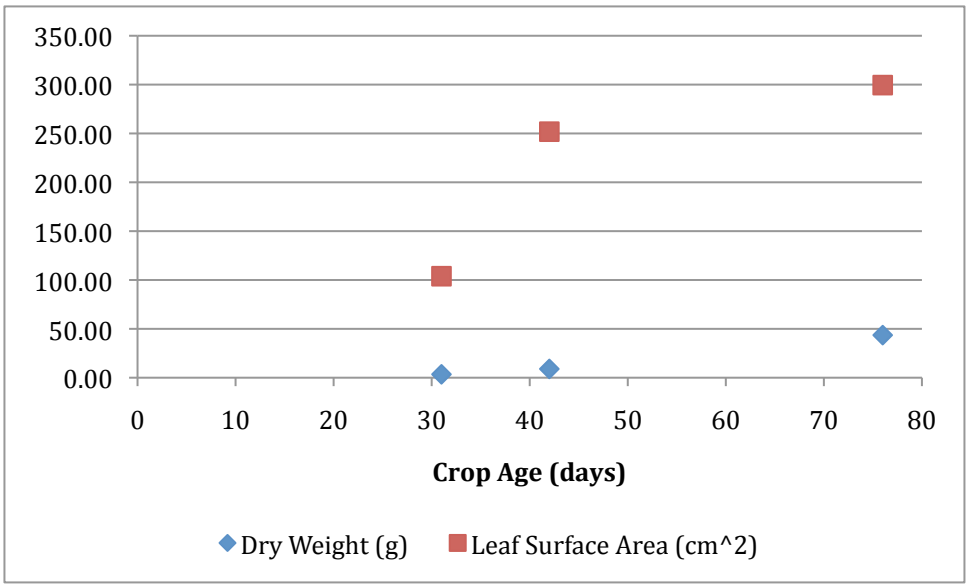

Figure 4-7 Trends in bean leaf area and standing biomass with crop development.

4.1.2.1 Effect of Plant Development

Table 4-3 breaks down the interception metric group averages and standard deviations for the three bean groups that received an identical volume of deposition at different stages of development.

Table 4-3 Bean interception results at varying stages of development.

Crop Age (days)		$f * 10^{-1} \pm \sigma$	$f_B * 10^{-1} \pm \sigma$	$f_{LAI} * 10^{-1} \pm \sigma$
A	31	1.2±.2	5.8±.66	1.9±.21
B	42	1.7±.32	3.2±.36	1.1±.12
D	76	2.6±.35	1.1±.22	1.5±.17

These results are visually presented in Figure 4-8. These are some of the most interesting results and the data is tightly clustered. The unitless interception fraction tends to increase as the plant develops, presumably as there is more surface area to retain intercepted activity. The mass interception fraction moves in the opposite direction with time, presumably as the increasing mass of bean pods makes standing biomass most likely to be surface-area-poor pod than leaves. The final metric f_{LAI} shows the least variability of the three, and there is not a general trend in its behavior. This is the ideal behavior for f_{LAI}, it is the closest thing to providing a development independant value for interception in beans.

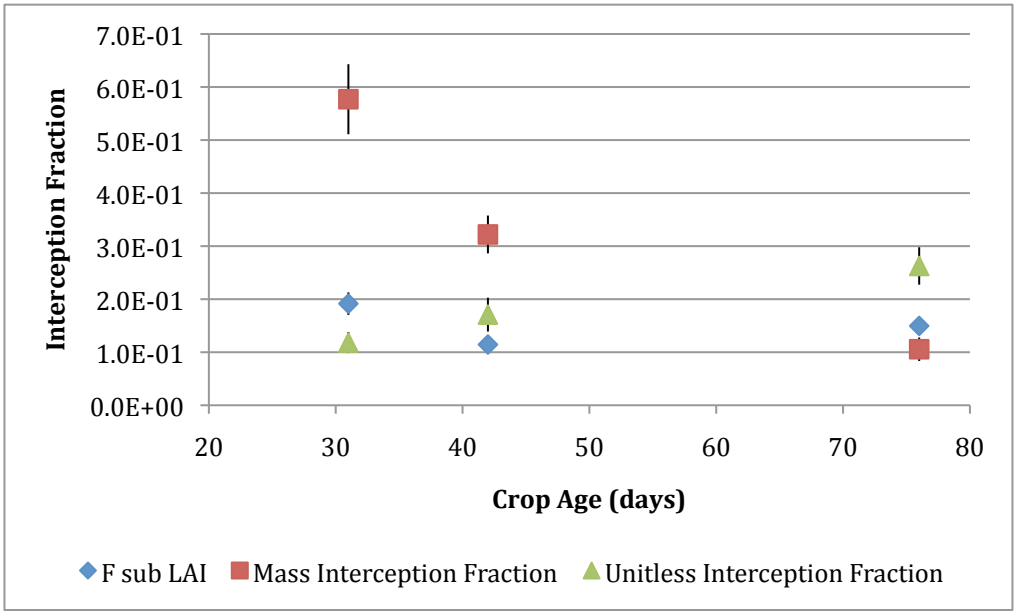

Figure 4-8 Interception fraction vs. crop age - bean groups A,B,D.

4.1.2.2 Effect of Deposition Volume

Table 4-4 breaks down the interception metric group averages and standard deviations for the five bean groups that received different volumes of deposition at a single stage of development.

Table 4-4 Bean interception results at varying deposition volumes.

Dep. Vol. (ml)		$f * 10^{-1} \pm\sigma$	$f_B * 10^{-1} \pm\sigma$	$f_{LAI} * 10^{-2} \pm\sigma$
C	17	3.9±.55	1.3±.28	18±1.7
D	34	2.6±.35	1.1±.22	15±1.7
E	68	2±.42	.63±.14	8.9±.88
D	136	1±.11	.45±.059	6.4±.49
G	272	.74±.074	.23±.052	3.3±.17

Again this shows the expected inverse relationship between intercepted activity and deposition volume. There does not seem to be significant adsorption to the bean leaves. The variability between metrics is again quite small, with all metrics decreasing by about a factor of 5.27, 5.65, and 5.45 respectively as the volume of deposition is increased by a factor of 16.

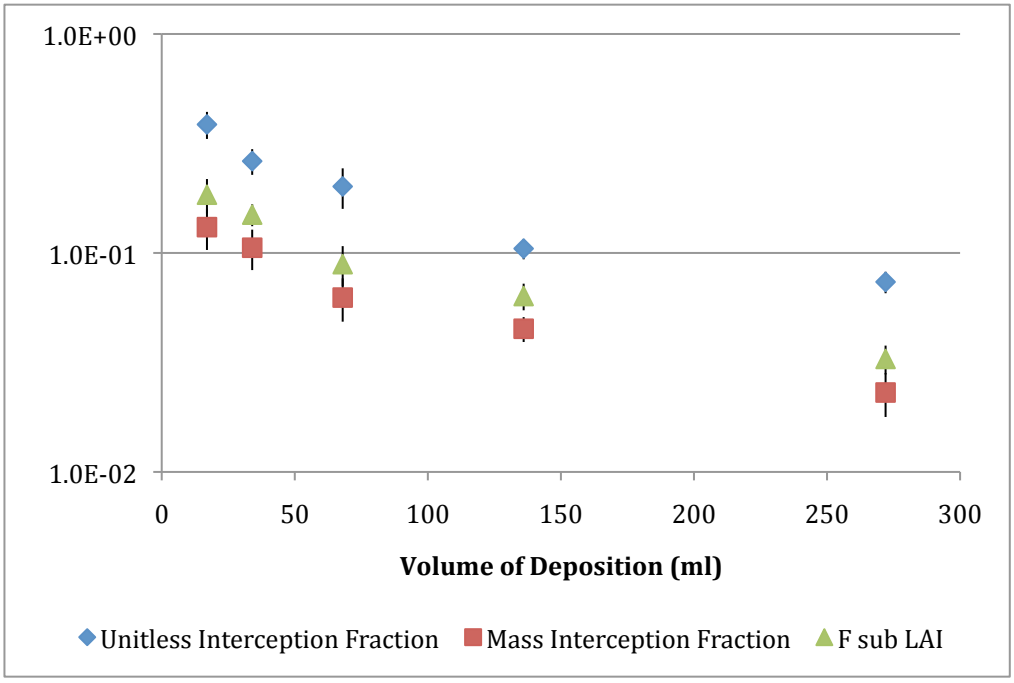

Figure 4-9 Interception fraction vs. deposition volume - bean groups C-G.

4.1.3 Wheat Interception Results

Figure 4-10 shows a much weaker correlation between leaf surface area and standing biomass than Figures 4-1 and 4-5. Wheat is the odd one out in a collection of radishes, beans, and wheat; its small bladed leaves have a tendency to shed water rather than retaining it. Interception group A is missing f_{LAI} results because it was initially decided that the LAI determination method was unsuitable for wheat due to differences in plant physiology. After some additional testing the decision was made to attempt measuring LAI for wheat although it was expected this metric would be less appropriate for wheat

than for leafy vegetables.

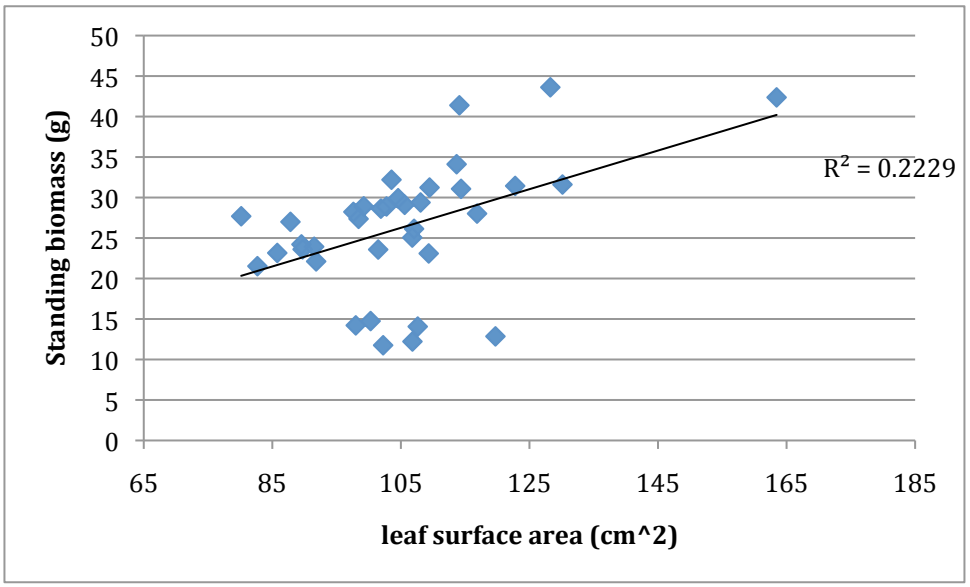

Figure 4-10 Leaf surface area vs. dry weight - all wheat data.

Figure 4-11 shows a history of leaf surfaces that is relatively flat while plant standing biomass increased as the crop developed.

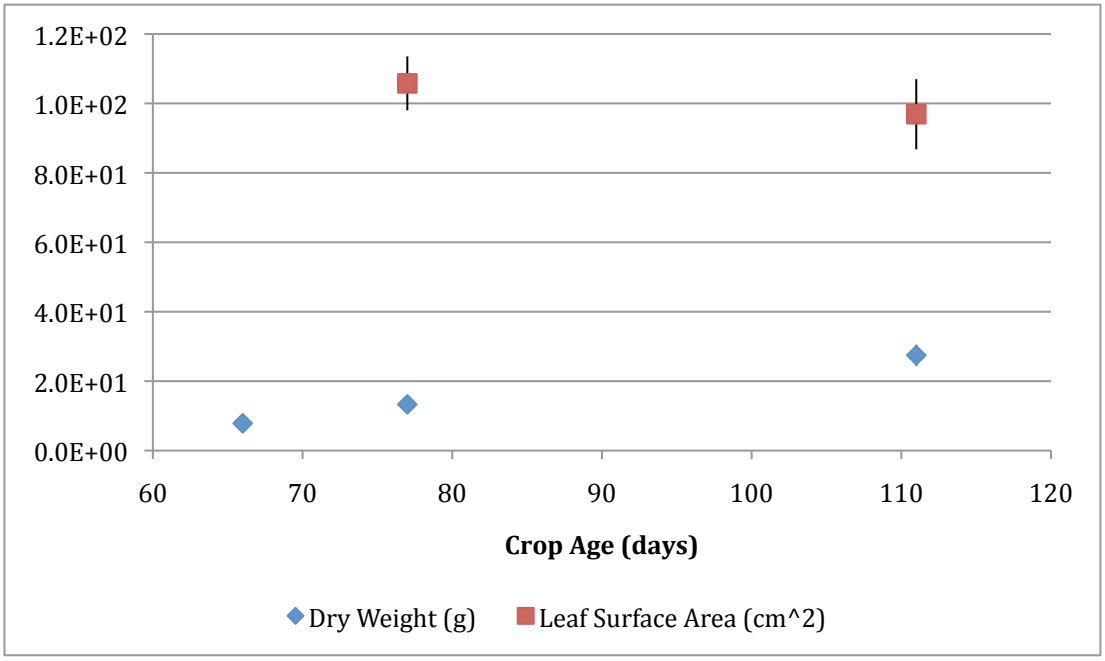

Figure 4-11 Trends in wheat leaf area and standing biomass with crop development.

4.1.3.1 Effect of Plant Development

Table 4-5 breaks down the interception metric group averages and standard deviations for the three wheat groups that received an identical volume of deposition at different stages of development.

Table 4-5 Wheat interception results at varying stages of development.

Crop Age (days)		$f * 10^{-1} \pm\sigma$	$f_B * 10^{-1} \pm\sigma$	$f_{LAI} * 10^{-1} \pm\sigma$
A	66	1.2±.25	2.6±.37	na
B	77	1.8±.65	2.3±.83	8.4±.12
D	111	3±.48	1.8±.22	6.5±.17

These results are visually presented in Figure 4-12. The unitless and mass interception fractions behave much as they did for bean plants; the unitless interception fraction increases with plant development while the mass interception fraction trails off. For the first time the mass interception fraction showed the least variability. The f_{LAI} data is difficult to interpret with two data points, but it does not seem to be as appropriate in wheat as it is in beans.

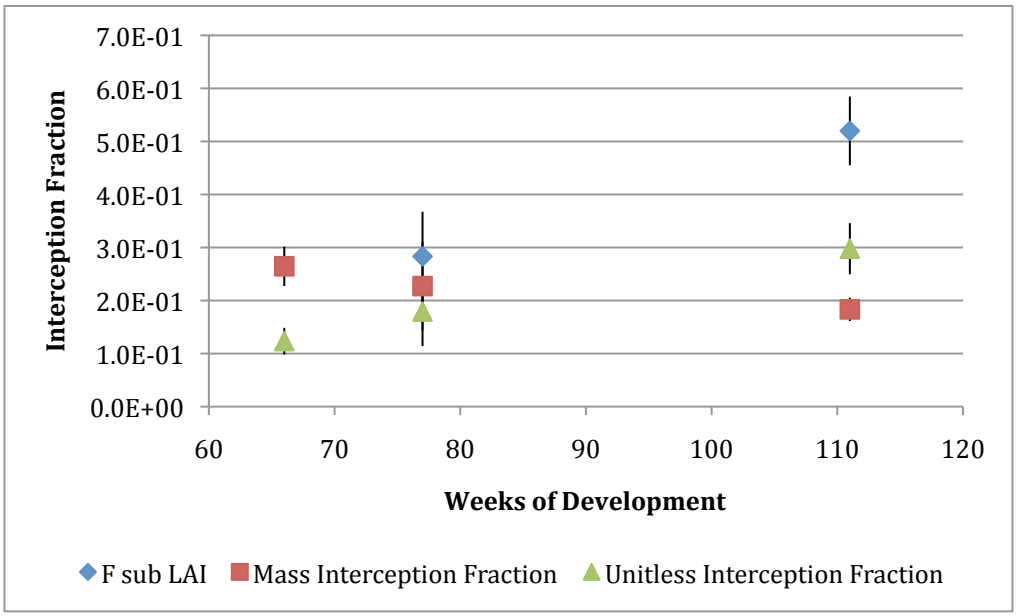

Figure 4-12 Interception fraction vs. crop age - wheat groups A,B,E.

4.1.3.2 Effect of Deposition Volume

Table 4-6 breaks down the interception metric group averages and standard deviations for the five wheat groups that received different volumes of deposition at a single stage of development.

Table 4-6 Wheat interception results at varying deposition volumes.

Dep. Vol. (ml)		$f * 10^{-1} \pm\sigma$	$f_B * 10^{-1} \pm\sigma$	$f_{LAI} * 10^{-1} \pm\sigma$
C	8.5	5.1±.76	3.1±.3	8.7±.66
D	17	4.8±.58	2.3±.46	6.7±.93
E	34	3±.48	1.8±.22	5.2±.65
D	68	2.1±.35	1.4±.13	3.4±.28
G	136	1.6±.35	.98±.12	2.6±.39

These data also show the expected inverse relationship between intercepted activity and deposition volume. The variability between metrics is again quite small, with all metrics decreasing by about a factor of 3.19, 3.16, and 3.35 respectively as the volume of deposition is increased by a factor of 16.

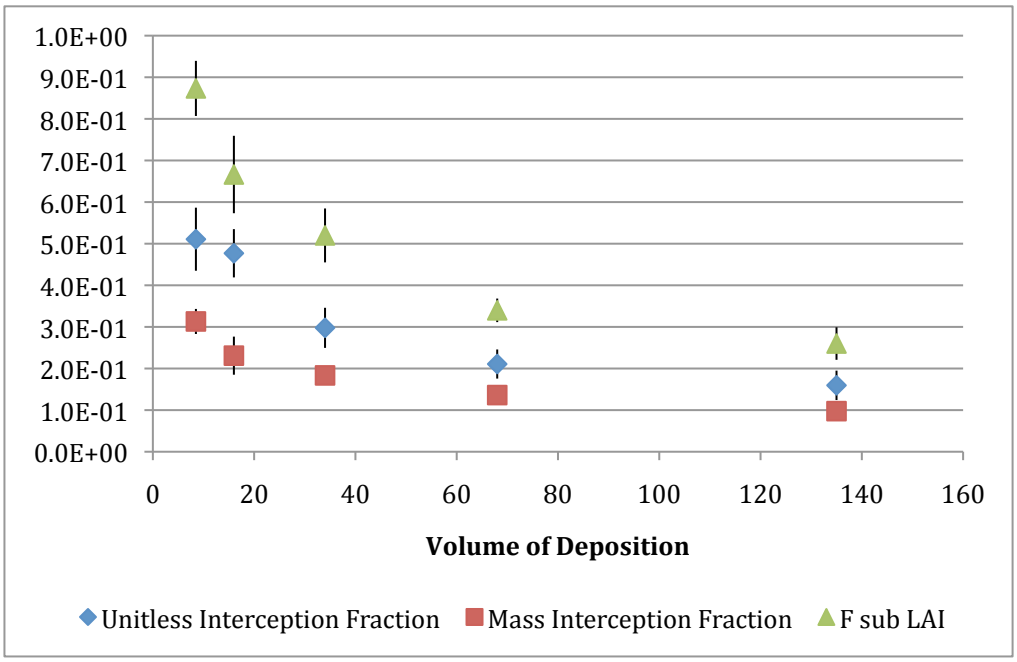

Figure 4-13 Interception fraction vs. deposition volume - wheat groups C-G.

4.1.4 Interspecies Interception Intercomparison

Table 4-7 has the range of interception metric values for each species from Tables 4-1, 4-3, and 4-5. These are numbers from plants at varying stages of development that received a volume of deposition approximately equivalent to 1mm rainfall.

Table 4-7 Range of interception metrics across species with constant deposition.

	f		**f_B**		**f_{LAI}**	
	min	max	min	max	min	max
Radishes	4.4e-02	6e-02	1.4e+00	4.6e+00	4.5e-02	1.4e-01
Beans	1.2e-01	2.6e-01	1.1e-1	5.8e-01	1.1e-01	1.9e-01
Wheat	1.2e-01	3.0e-01	1.8e-01	2.6e-01	2.8e-01	5.2e-01

The variation between crops is fairly small, less than an order of magnitude for f and f_{LAI} and only slightly greater than that for f_B.

4.2 Foliar Absorption and Translocation

4.2.1 Radish Absorption Results

Table 4-8 breaks down the absorption metric group averages and standard deviations for the four radish groups that were harvest a variable period of time after a foliar deposition.

Table 4-8 Radish absorption results at 4 different times after ^{36}Cl application.

Days after Treatment		ROR (in % ±σ)	ROA (in % ±σ)	IA (in % ±σ)
A	1	94±2	86.8±3	94.4±2.1
B	2	97.1±1.1	82.6±12.1	97.6±0.6
C	4	97.7±.8	92.2±7.8	97.8±0.9
D	10	96.8±1	92.2±8.3	96.9±0.9

These results will set the pattern for the remainder of the absorption experiments. Plants quickly and almost completely absorbed foliar deposited ^{36}Cl. There appears to be a slight increase after 24 hours in the ROR and IA methods, but not in the ROA method. A high value was expected and all three metrics report similar values. The ROR method measures absorption by taking the amount recovered in the plant and dividing it by the total activity recovered in the plant and wash water used to wash the treated leaves. Its results agree very closely with the IA method, which calculates absorption by taking the activity applied, subtracting the activity recovered in the wash

water, and dividing the result by the activity applied. These two methods are quite similar and have the virtue of being unable to produce absorption values greater than 100%. The ROA method is the total activity recovered from the plant divided by the activity deposited. The ROA method's predictions show significantly more variability: this could be attributable to imprecision in radionuclide application.

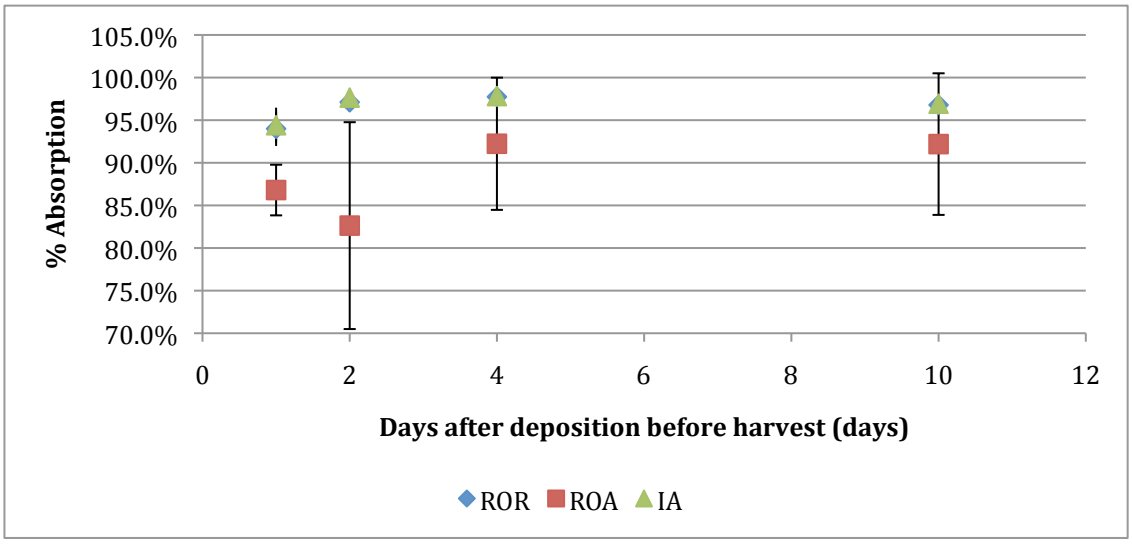

Figure 4-14 Absorption vs. time before harvest in radishes.

The absorption experiments also allowed for a mass balance tracking the fate of the ^{36}Cl in the system using the known deposition, the activity concentrations in edible and crop and foliage, and the masses of each compartment. In the radish experiments the total recovery was high, a certain loss is expected and could be attributable to a number of circumstances, e.g. exudation through the roots or loss to insect ingestion. The activity recovered from the wash water decreased after the first treatment group, indicating absorption quickly reached a maximum. A slight increase in the final group could be a result of partial leaf senescence.

Table 4-9 Radish absorption experiments mass balance.

	Activity in plant (%)	Activity in radish (%)	Activity in wash water (%)
A	84	3	6
B	77	5	2
C	89	3	2
D	83	9	3

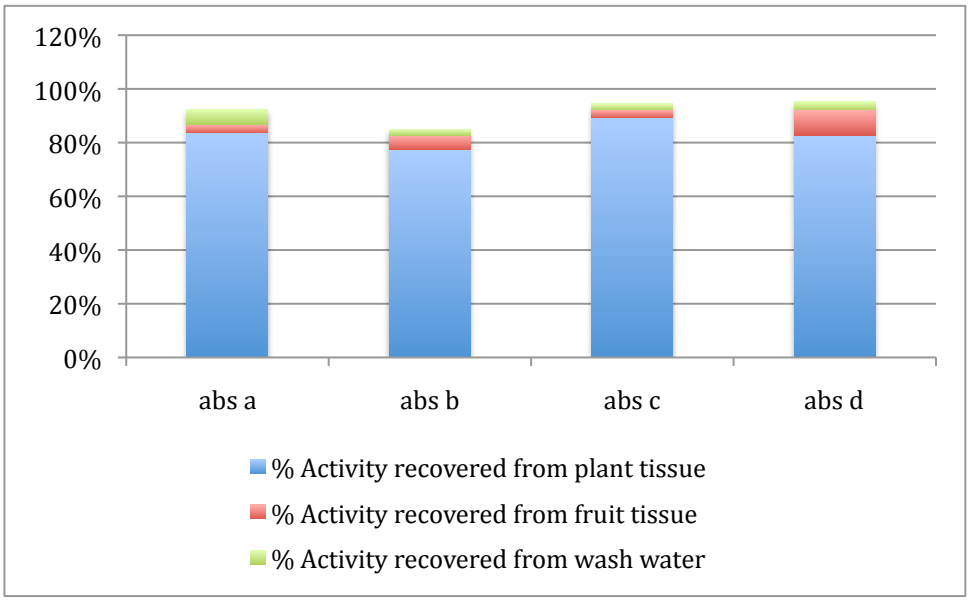

Figure 4-15 Total recovery in radish absorption experiments.

4.2.2 Radish Translocation Results

Table 4-10 breaks down the translocation metric group averages and standard deviations for the three radish groups that were treated with ^{36}Cl a variable period of time before harvest.

Table 4-10 Radish translocation results.

Crop age at treatment		ROR (in % ±σ)	ROA (in % ±σ)	CR (in % ±σ)
A	2	5±3	4±3	4±2
B	4	11±4	11±4	6±2
C	30	15±3	17±3	8±1

The ROR and ROA method show very good agreement here, barely differing. The CR method shows less translocation than the others, this is attributed to the fact that this metric does not take into account the relative mass of the edible radish compared with rest of the plant. TLOC B was treat the same day as ABS A,B,C, and D and since ABS D is basically the same experiment as TLOC B except that the treated leaves of ABS D were washed before analysis. The translocated fractions in ABS D and TLOC B are 9% and 11% respectively, within one standard deviation. Translocation in radishes continued to increase until harvest, pouring increasing resources in the edible radish. This is consistent with the observations, that leaf number and sizes remained constant after the first couple weeks after which the only visible changes were an increase in the size of the edible radish visible at the base of the plant.

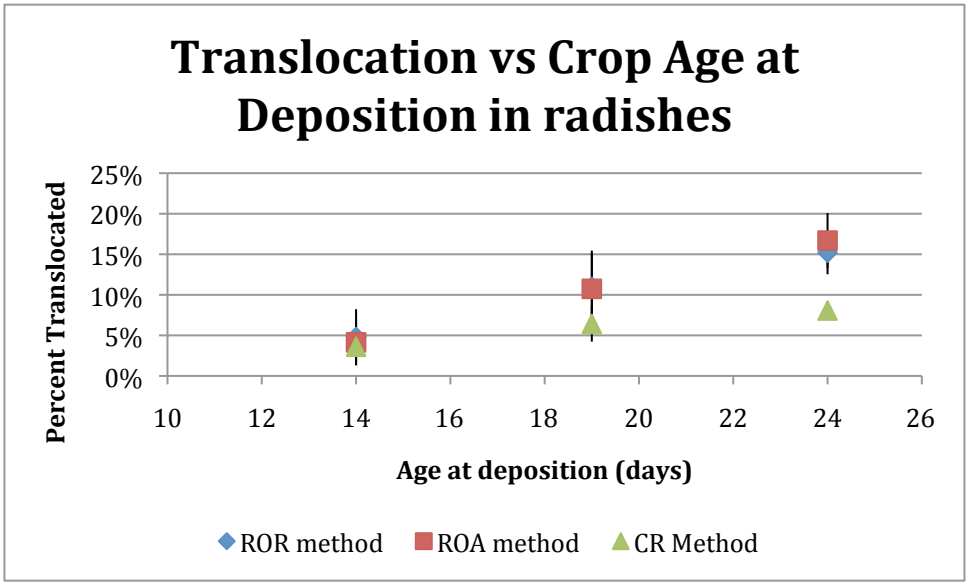

Figure 4-16 Translocation vs. crop age at deposition in radishes.

The range in activity recovery was similar to the radish absorption experiments. Recovery was higher in the experiments were less time elapsed between treatment and harvest, this is not unexpected as the more time passes; the more there is potential for loss. TLOC C actually reports recovery of greater than 100%, this is of course not physically the case. Potential causes for this result are discussed in more detail in the wheat translocation section.

Table 4-11 Radish translocation experiments mass balance.

	Activity in plant (%)	Activity in radish (%)
A	85	4
B	87	11
C	93	17

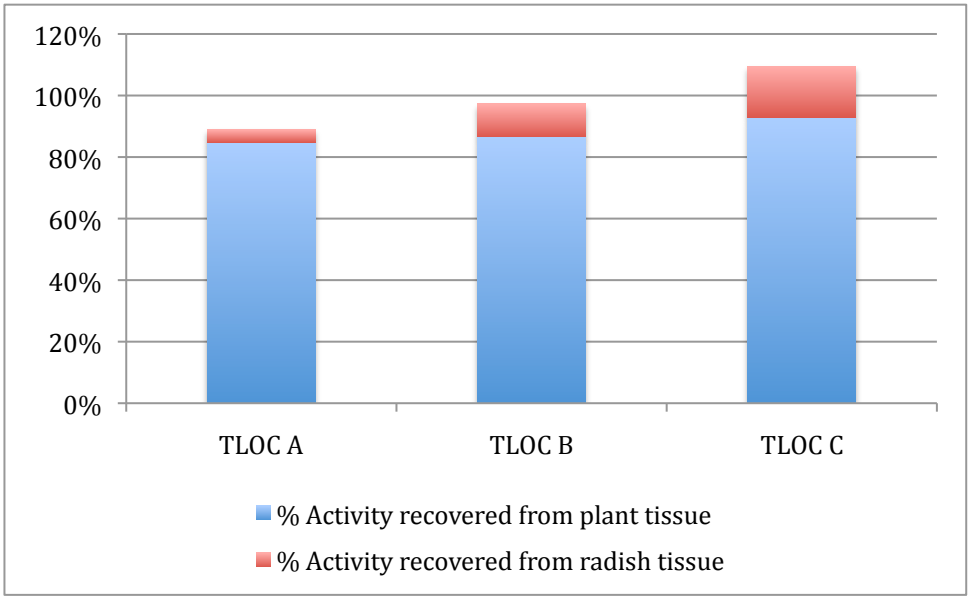

Figure 4-17 Mass balance for radish translocation experiments.

4.2.3 Bean Absorption Results

Table 4-12 breaks down the absorption metric group averages and standard deviations for the three bean groups that were harvest a variable period of time after a foliar deposition.

Table 4-12 Bean absorption results at three different times after ^{36}Cl application.

Days after Treatment		ROR (in % ±σ)	ROA (in % ±σ)	IA (in % ±σ)
A	2	97±1	88±17	98±1
B	4	98±0	98±16	98±0
C	30	93±4	85±14	94±3

The first two results agree closely with the radish experiments, showing very quick uptake. The results for the third group are contrary to this trend. The suspected reason for the decrease is that at the time of harvest some of the early leaves had begun to senesce; the wash water used to wash ABS C was considerably darker than that for ABS B or C. It is believed this hue is due to breaking down of dead contaminated leaf tissue in the wash water releasing ^{36}Cl and depressing the ROR and IA metrics.

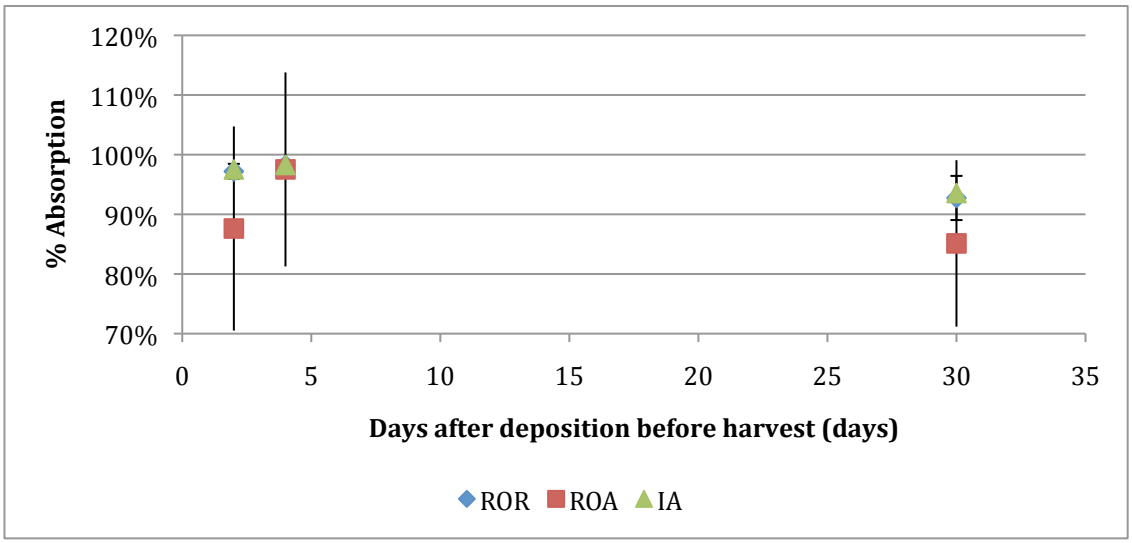

Figure 4-18 Absorption vs. time before harvest in beans.

Table 4-13 has the mass balance data for the bean absorption experiments. Again the total recovery was high. The activity recovered from the wash water remained practically steady for the first two treatment groups, indicating absorption quickly reached a maximum. The increase in the activity in the wash water in the final group was attributed to the senescence of treated leaf tissue and the breakdown of the leaf tissue in the washing process. The 10% translocation to the bean pod in ABS C

matches well with the 9% - 10% translocation seen in TLOC C (see next section), as with radish groups ABS D and TLOC B these experiments were coincident and the equality of outcomes here is reassuring.

Table 4-13 Bean absorption experiments mass balance.

	Activity in plant (%)	Activity in bean pod (%)	Activity in wash water (%)
A	85	3	2
B	94	3	2
C	75	10	6

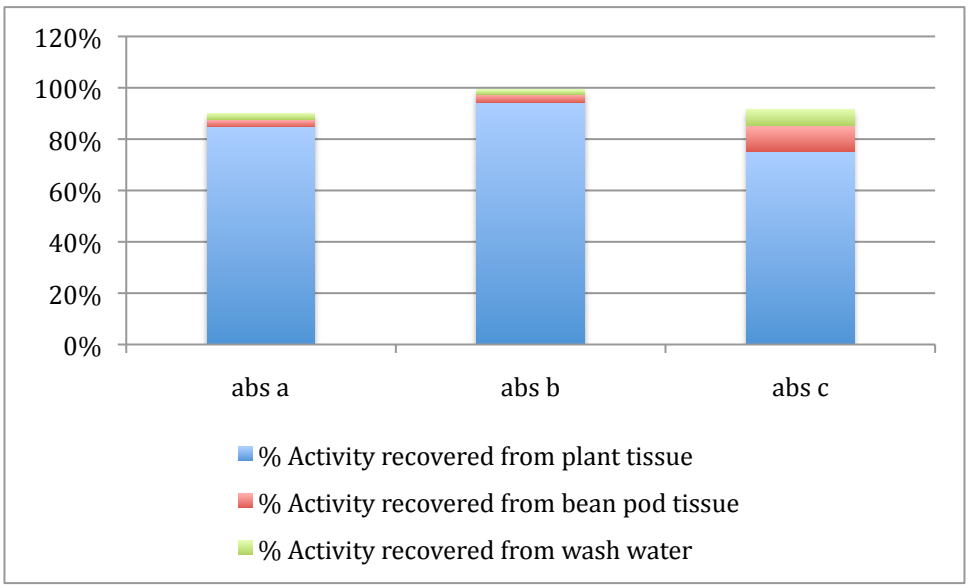

Figure 4-19 Mass balance for bean absorption experiments.

4.2.4 Bean Translocation Results

Table 4-14 breaks down the translocation metric group averages and standard deviations for the five bean groups that were treated with ^{36}Cl a variable period of time before harvest.

Table 4-14 Bean translocation results after ^{36}Cl application at different stages.

Crop age at treatment		ROR (in % ±σ)	ROA (in % ±σ)	CR (in % ±σ)
A	4	9±2	8±2	5±1
B	6	15±2	16±4	9±2
C	8	10±3	9±2	5±1
D	10	3±1	3±1	2±1
E	12	1±.1	1±.1	0±.1

The ROR and ROA method again show a very close agreement while the CR method reports significantly less translocation than the other two metrics. Between translocation and absorption, translocation seems to be a more consistent process. The pattern of translocation's relationship to age at treatment is different to that of radishes, translocations rises, but then steadily decreases. This behavior is attributed to the plant's physiology; the experiments were begun at the time of first flowering, before that time there was no bean pod to accumulate any activity. The fraction translocated peaks at six weeks. Presumably this is when the plant is most focused on its developing pods. After that the translocated fraction steadily decreases, the bean pods are increasingly mature requiring fewer nutrients from the plant and there is less time before harvest in which translocation could occur.

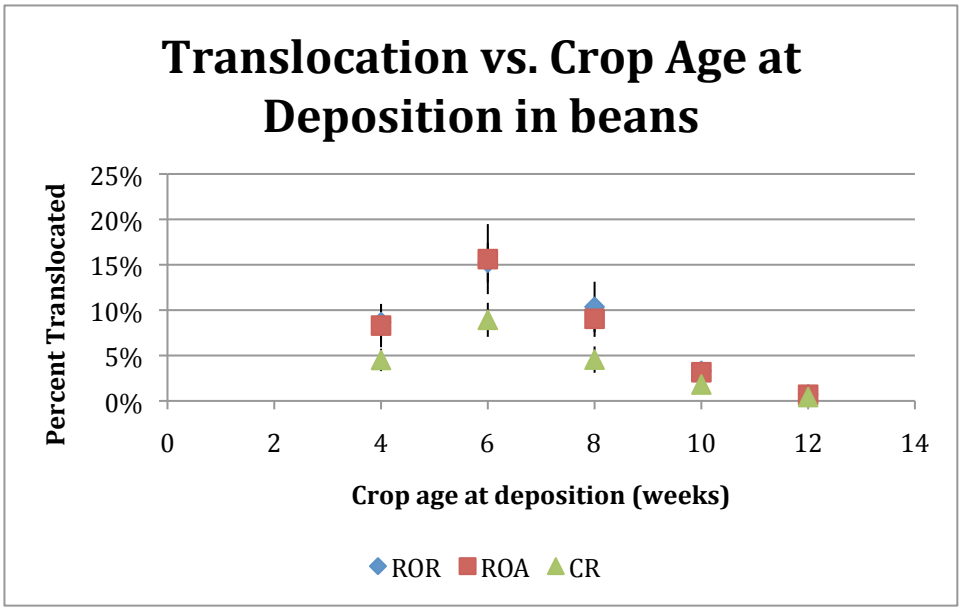

Figure 4-20 Translocation vs. crop age at deposition in beans.

The mass balance for the bean translocation experiments is similar to the previous balances. The recovery is high and shows no real trend over the course of the experiments.

Table 4-15 Bean translocation experiments mass balance.

	Activity in plant (%)	Activity in bean pod (%)
A	87	8
B	86	16
C	80	9
D	92	3
E	94	1

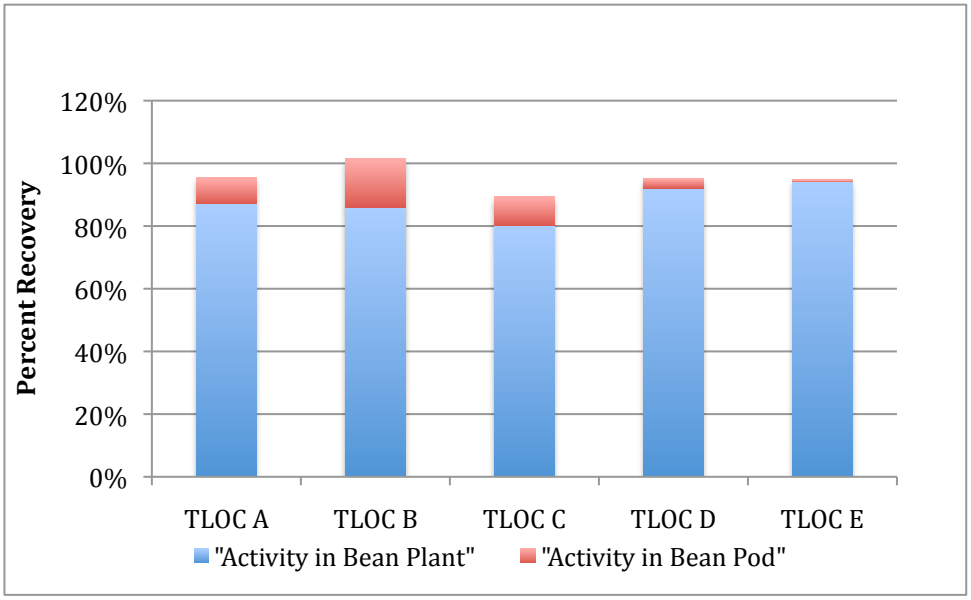

Figure 4-21 Mass balance for bean translocation experiments.

4.2.5 Wheat Absorption Results

Table 4-16 breaks down the absorption metric group averages and standard deviations for the three wheat groups that were harvest a variable period of time after a foliar deposition.

Table 4-16 Wheat absorption results at three different times after ^{36}Cl application.

Days after Treatment		ROR (in % ±σ)	ROA (in % ±σ)	IA (in % ±σ)
A	2	97±1	114±10	97±1
B	4	98±1	112±8	98±1
C	30	94±3	115±13	93±3

The results are similar to the crops above; overall absorption is very high. As usual the ROR and IA method agree very closely while the ROA numbers differ considerable.

The ROA numbers are in excess of one, which is not reasonable. Similarly high numbers will be seen in the total recovery of the wheat translocation experiments and the matter is discussed in more detail in Section 4.2.6. As with the beans, the decrease in the ROR and IA results for ABS C is attributed to dead leaf tissue breaking up in the wash water and releasing previously absorbed radionuclide. At the time of harvest many of the early leaf blades had begun to senesce.

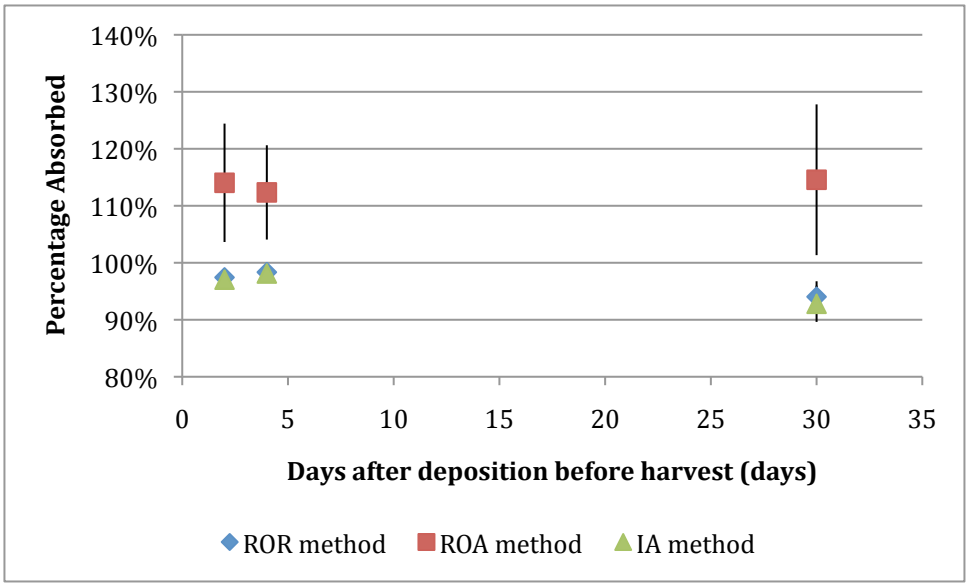

Figure 4-22 Absorption vs. time before harvest in wheat.

The mass balance issues are similar for both the wheat absorption and translocation experiments and are discussed in the next section.

Table 4-17 Wheat absorption experiments mass balance.

	Activity in plant (%)	Activity in grain (%)	Activity in wash water (%)
A	110	4	3
B	107	6	2
C	87	27	7

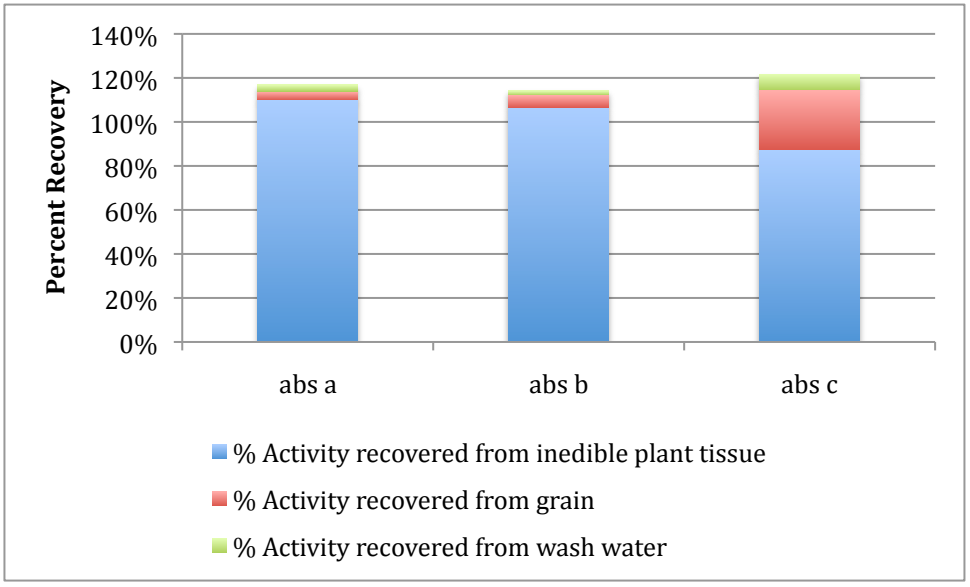

Figure 4-23 Mass balance for wheat absorption experiments.

4.2.6 Wheat Translocation Results

Table 4-13 breaks down the translocation metric group averages and standard deviations for the five wheat groups that were treated with ^{36}Cl a variable period of time before harvest.

Table 4-18 Wheat translocation results after ^{36}Cl application at different stages.

Crop age at treatment		ROR (in % ±σ)	ROA (in % ±σ)	CR (in % ±σ)
A	4	30±7	33±9	31±11
B	6	32±2	40±12	29±10
C	8	21±5	24±6	16±6
D	10	19±4	20±4	15±5
E	12	9±3	11±4	7±3

The wheat shows the same pattern of translocation as the beans did. The differences are minor, but for the first time CR method trends differently than the other two metrics. The CR method reports steadily declining translocation while the other two report an increase in the second group. As with the beans, experiments were timed to begin at approximately the time of flowering. Translocation initially rose to a peak, and then steadily declined, presumably for the same reasons as in the bean translocation experiments. The translocation numbers are significantly higher for wheat than for either of the other crops.

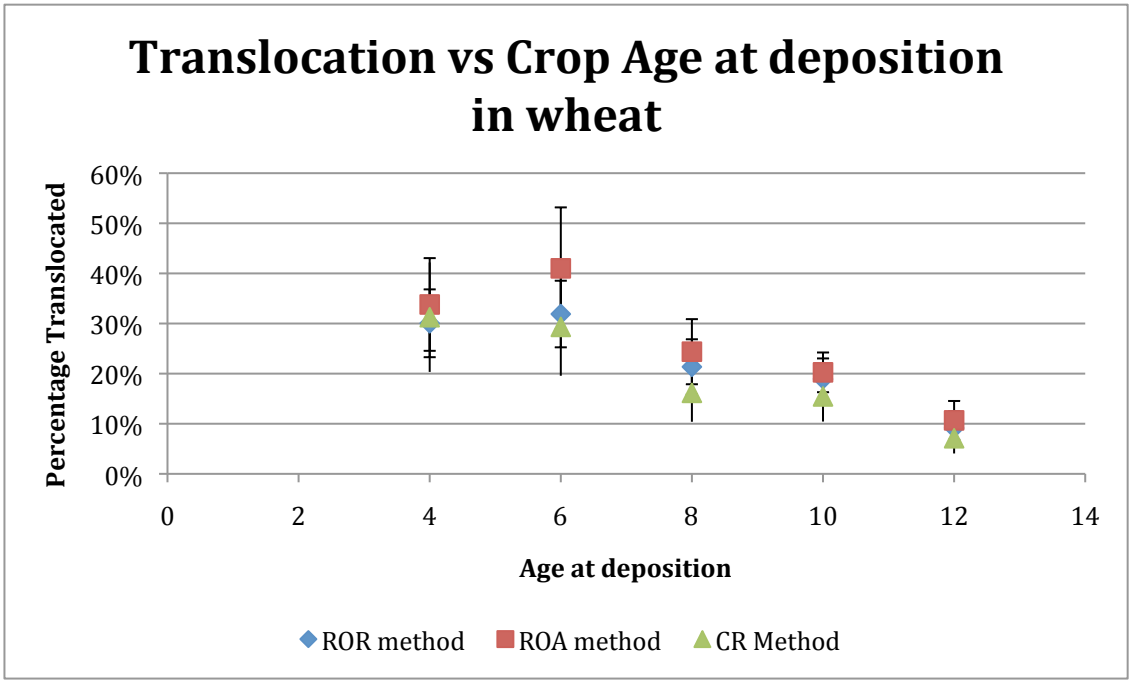

Figure 4-24 Translocation vs. crop age at deposition in wheat.

While sporadic in the case for radishes and beans, in wheat the reported recovery was consistently in excess of 100%, as shown in table 4-19. A variety of explanations for this problem were considered, chief among them; flaws in the plant activity determination process, systematic bias in equipment such as scales or pipettes, and overdispensing of contaminating solution onto the wheat plants. The first explanation considered was to examine flaws in the plant activity determination process. Radiation Center Radiation Safety made a Gas Flow Proportional Counter available for this purpose. This gas flow proportional counter was used earlier in this work with a sealed ^{36}Cl standard to verify the ability of the LSC to detect ^{36}Cl in water at essentially 100% efficiency. In the earlier work the GFPC agreed with the LSC to within 4%. To evaluate the plant activity determination process 0.25 g of wheat samples TLOC A3

and TLOC C5 were taken from the excess plant material retained after analysis via LSC. These samples were placed on a planchet and fixed into place with hair apray.

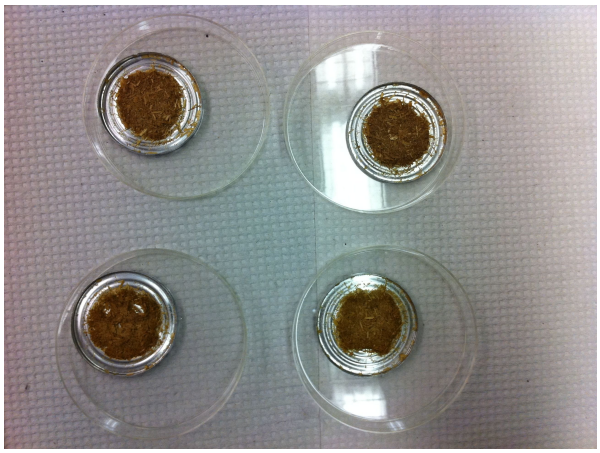

Figure 4-25 Wheat samples prepared for analysis via GFPC.

According to the LSC analysis these samples (Samples A and B) had a dry weight activity of 263.7 Bq g^{-1} and 228.0 Bq g^{-1} respectively so the activity the samples were expected to contain was 65.9 Bq and 57 Bq of ^{36}Cl respectively. Two other samples (Samples C and D) were prepared by taking 0.25 g of uncontaminated wheat tissue and adding 65.9 Bq and 57 Bq of ^{36}Cl respectively, these samples were also fixed in place with hair spray. The samples were each counted four times for 10 minutes apiece in order from A-D, then they were counted again in the same way. The results of the second set of 16 ten minute counts is included below as Table 4-19.

Table 4-19 GFPC results to validate plant activity determination procedure.

Crop age at treatment	Count 1 (CPM)	Count 2 (CPM)	Count 3 (CPM)	Count 4 (CPM)	Average ± Std Dev.
A	980.0	1008.3	1037.0	1071.5	1024.2±39.2
B	699.5	744.3	740.0	749.5	733.3±22.9
C	988.5	1027.0	1039.8	1074.3	1032.4±35.4
D	705.5	737.8	730.5	767.8	735.4±25.6

The ratios of the averages of A/C and B/D respectively are .997 and .976, showing an extremely close agreement. This rules out most potential flaws in the plant activity determination method. It is still possible that there were heterogeneities in the blended plant samples where portions of the plant material with more or less activity was lost during the blending process, but that is considered unlikely. The close agreement between the LSC and the GFPC also rules out differences in behavior during digestion between spiked blanks and true samples as a cause for the high recovery percentages.

The balance used in these experiments was tested using standard masses and not found to have a bias. The pipettes were tested gravimetrically on the calibrated balance and again no bias was discovered. The remaining explanation of an over application of ^{36}Cl

remains the most likely. Potentially human error such as tilting the pipettes in awkward directions could have led to additional unwanted deposition. ^{36}Cl was applied to the wheat in twenty-four 25uL drops where the drops were 100uL for beans. It is possible that this large number of applications could have carried extra ^{36}Cl if the pipette was tilted as was sometimes necessary to access the appropriate area of the plant. No completely satisfactory explanation was found for these implausibly high recovery values. The precision of most of the treatment groups and the agreement of the GFPC and the LSC contribute to confidence in the results, but the high recovery numbers remain a concern. However while remaining a matter of concern, they have relatively little influence on most of the results reported here. Most of the metrics compare the activities in different plant compartments and any overestimation cancels out. In choosing conservative parameters for radioactive waste modeling it would be prudent to choose the maximum translocation or interception value at the time of peak translocation or interception and in comparison the excess reported is insignificant even in metrics where it would alter the numbers.

Table 4-20 Wheat translocation experiments mass balance.

	Activity in plant (%)	Activity in grain (%)
A	76	33
B	84	40
C	89	24
D	86	20
E	106	11

It was interesting to note that mass balance data is rarely presented in peer reviewed articles and many of the metrics that are used are designed in such a way that it is impossible for results such as greater than 100% absorption to be reported.

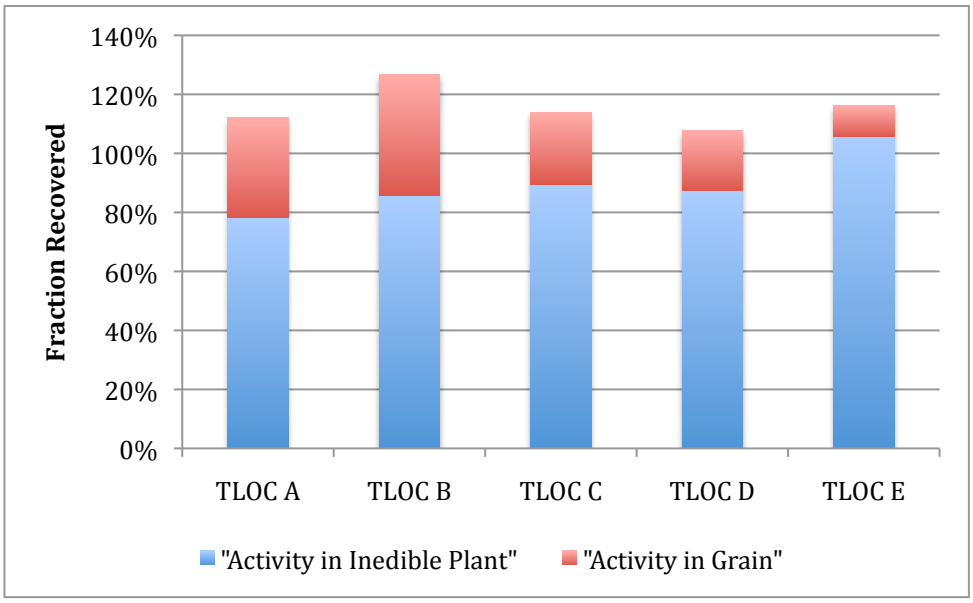

Figure 4-26 Mass balance for wheat translocation experiments.

4.2.7 Interspecies Absorption and Translocation Results

Table 4-21 has the range of absorption metric values for each species from Tables 4-8, 4-12, and 4-16.

Table 4-21 Range of absorption metrics across species.

	ROR (%)		ROA (%)		IA (%)	
	min	max	min	max	min	max
Radishes	94	97	82	92	94	98
Beans	93	98	85	98	94	98
Wheat	94	97	112	115	93	97

The agreement between metrics is strong, with the ROA method providing outlier data as usual. The difference between plant species seems minimal as well.

Table 4-22 has the range of translocation metric values for each species from Tables 4-10, 4-14, and 4-18.

Table 4-22 Range of translocation metrics across species.

	ROR (%)		ROA (%)		CR (%)	
	min	max	min	max	min	max
Radishes	5	15	4	17	4	8
Beans	1	15	1	16	2	5
Wheat	9	30	11	34	7	31

The agreement between methods is strong, with the CR method generally reporting lower values, but species pays a much larger role than in absorption.

5 Conclusion

Crop interception of ^{36}Cl is found to generally follow the patterns that were expected and discussed in the literature review: with increasing deposition there is decreased retention. Sporadic irrigation will result in significantly greater interception than a similar volume of continuous irrigation. For this reason it is emphasized that choosing appropriate irrigation volumes per deposition event is key in parameter selection. Many current models do not spend much or any time discussing this matter and the lack of this discussion is a weakness in the safety case. The role of plant development is also important; in beans and wheat where the fruiting body is aboveground unitless interception fractions tend to increase as standing biomass increases. There is a trend in the opposite direction for the mass interception fraction as the plant develops. This trend is not present in radishes where most of the growth in plant mass later in the crop's life is below ground and not captured by either LAI or standing biomass. For that reason the unitless interception fraction showed the least variation in radishes, though variation in all the metrics was reduced as compared to beans. For beans the use of f_{LAI} was found to show the least dependence on plant development. For leafy green vegetables where the fruiting body is aboveground, the f_{LAI} was the superior metric. It was found to be less useful in wheat; where the mass interception fraction showed the least variability. This is attributed to the significant physiological differences between the leaves of beans and radishes and those of wheat. This difference is illustrated by the very different relationship between LAI and standing biomass in radishes and beans as

compared to wheat. The lesson from the investigations into the effect of plant development on interception metrics is that the physiology of the plants involved plays a significant role, no one metric is superior in all situations. The decision in current models in the United States to use the unitless interception fraction is a reasonable one.

Foliar absorption of ^{36}Cl was found to approach 100%. The ROR and IA absorption methods produced nearly identical results, with very little intragroup variation, in contrast with the ROA absorption method that showed more variability between and within groups. The IA method is substantially easier to perform since it does not require the analysis of plant tissue and may be the best metric for quantifying absorption for that reason. No matter which metric was used, all crops were found to absorb the bulk of the ^{36}Cl deposited on their leaves in short order. This is not surprising when the crop science literature is consulted, but differs significantly from the values in use in radiological assessment. Recent repository assessments use a 50% absorption rate that significantly underestimates the potential for foliar uptake and translocation (EPRI 2009). While some of the results of this work may not be applicable under different environmental conditions, the magnitude of foliar absorption should be constant in a wide variety of environments and it would be prudent for future assessments to assume 100% absorption of intercepted ^{36}Cl. This has a side effect of making weathering terms relatively unimportant for ^{36}Cl unless the weathering occurs very swiftly after deposition. The volume of deposition still plays a key role.

The radiological community could benefit from increased collaboration with crop and soil scientists. The plant science community has a better understanding of the physiological processes involved and their methods report higher translocation in nearly all cases. The ROA and ROR translocation methods look at how much activity was deposited and where it went, whereas the CR method propounded in the IAEA's recent technical report looks at the activity concentrations between edible plant and foliage, ignoring the mass of each compartment. In addition to making more sense, the ROR and ROA translocation metrics show a very close agreement while the CR method reports consistently lower values for translocation. Translocation metrics showed much more variation within species than the absorption metrics; radishes and beans had relatively similar translocation values, but wheat show significantly greater translocation to the fruiting body.

The results presented here provide data for ^{36}Cl that do not exist in the literature, yet perhaps the most useful results of this work are the qualitative comparisons of the behavior of many different metrics related to foliar interception, absorption, and translocation, many of which have not appeared in the radiological literature before. The safe disposal of radioactive wastes depends on understanding the systems involved as well as possible and several of the foliar uptake metrics introduced here are improvements on those currently in use in radiological assessment. Biosphere models are the step immediately preceding uptake by people in performance assessment; a defensible safety case for the responsible disposal of radioactive waste depends on

identifying and addressing gaps in the literature and making use of the best science to explain the choices of model and parameter made in assessing potential risks to human beings.

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A Tables of Results

This table has data for each plant in the study. Each table includes the raw count rate for each sample, quantities derived from that raw count rate, and numbers relevant to the experiments such as activity deposited. The process at arriving at activity from count rate is as follows: A background count rate is subtracted from sample count rate. This corrected count rate can be thought of as having units of counts per minute per 0.1 g dried sample. Divide the result by 60 to express it in "per second" terms, then multiply by 10 to change the basis to "per gram." Multiplying by the appropriate correction factor in table A-1 results in a measurement of radioactivity: Bq g^{-1} (dried tissue). All of the numbers needed to calculate any of the metrics reported above are included in this appendix. Further rationale for the efficiency correction factor is found in the main body of the document.

	Inedible plant	Edible plant	Whole plant
Radishes	1.6e+00	1.5e+00	1.5e+00
Beans	1.7e+00	1.5e+00	1.7e+00
Wheat	2.1e+00	1.8e+00	2.0e+00

Table A-1 Efficiency corrections factors.

A.1 Radish Results

The averaged background count rate (CPM) for each type of radish sample, radish, plant tissue, and total plant were respectively 16.2, 16.3, 16.25.

A.1.1 Radish Interception Results

Sample	Count rate (CPM)	Plant dry weight (g)	Plant Activity, (Bq g^{-1})	Leaf area, (cm^2)

A1	4136	0.121	1.07E+03	4.4E+01
A2	2696	0.149	6.94E+02	4.8E+01
A3	3304	0.136	8.51E+02	4.4E+01
A4	2591	0.132	6.66E+02	4.3E+01
A5	3003	0.123	7.73E+02	3.7E+01
A6	2556	0.131	6.57E+02	4.2E+01
B1	1215	0.325	3.10E+02	9.9E+01
B2	1820	0.297	4.67E+02	8.2E+01
B3	1996	0.244	5.12E+02	7.3E+01
B4	1228	0.233	3.14E+02	8.3E+01
B5	1303	0.207	3.33E+02	7.3E+01
B6	1412	0.251	3.61E+02	7.1E+01
C1	1528	0.291	3.91E+02	9.8E+01
C2	788	0.258	2.00E+02	8.9E+01
C3	1009	0.305	2.57E+02	1.0E+02
C4	1021	0.417	2.60E+02	1.1E+02
C5	828	0.275	2.10E+02	8.3E+01
C6	608	0.307	1.53E+02	1.1E+02
D1	1548	0.407	3.96E+02	1.2E+02
D2	2493	0.177	6.41E+02	6.0E+01
D3	1067	0.336	2.72E+02	1.1E+02
D4	1638	0.364	4.20E+02	1.0E+02
D5	708	0.377	1.79E+02	8.4E+01
D6	2288	0.302	5.88E+02	9.6E+01
E1	4003	0.298	1.03E+03	8.9E+01
E2	1808	0.231	4.64E+02	6.4E+01
E3	13016	0.308	3.36E+03	1.0E+02
E4	2537	0.328	6.52E+02	1.0E+02
E5	7892	0.223	2.04E+03	8.2E+01
E6	1426	0.326	3.65E+02	1.1E+02
F1	2103	0.37	5.40E+02	9.8E+01
F2	2000	0.31	5.13E+02	1.1E+02
F3	15848	0.28	4.10E+03	9.5E+01
F4	9302	0.267	2.40E+03	8.4E+01
F5	11815	0.345	3.05E+03	1.0E+02
F6	7791	0.328	2.01E+03	1.0E+02
G1	946	0.278	2.41E+02	9.2E+01
G2	885	0.399	2.25E+02	8.8E+01
G3	1153	0.331	2.94E+02	1.1E+02

G4	1356	0.287	3.47E+02	9.2E+01
G5	1089	0.358	2.78E+02	9.6E+01
G6	1471	0.209	3.76E+02	5.8E+01

Table A-2 Radish interception results.

Sample	**Activity Deposited (Bq)**	**f**	**f_B**	**f_{LAI}**
A1	1.72E+03	7.5E-02	6.2E+00	1.7E-01
A2	1.72E+03	6.0E-02	4.0E+00	1.2E-01
A3	1.72E+03	6.7E-02	4.9E+00	1.5E-01
A4	1.72E+03	5.1E-02	3.9E+00	1.2E-01
A5	1.72E+03	5.5E-02	4.5E+00	1.5E-01
A6	1.72E+03	5.0E-02	3.8E+00	1.2E-01
B1	1.72E+03	5.8E-02	1.8E+00	5.9E-02
B2	1.72E+03	8.0E-02	2.7E+00	9.8E-02
B3	1.72E+03	7.3E-02	3.0E+00	9.9E-02
B4	1.72E+03	4.2E-02	1.8E+00	5.1E-02
B5	1.72E+03	4.0E-02	1.9E+00	5.4E-02
B6	1.72E+03	5.3E-02	2.1E+00	7.4E-02
C1	1.72E+03	6.6E-02	2.3E+00	6.8E-02
C2	1.72E+03	3.0E-02	1.2E+00	3.3E-02
C3	1.72E+03	4.5E-02	1.5E+00	4.5E-02
C4	1.72E+03	6.3E-02	1.5E+00	5.7E-02
C5	1.72E+03	3.4E-02	1.2E+00	4.1E-02
C6	1.72E+03	2.7E-02	8.9E-01	2.5E-02
D1	3.45E+03	4.7E-02	1.1E+00	4.1E-02
D2	3.45E+03	3.3E-02	1.9E+00	5.4E-02
D3	3.45E+03	2.6E-02	7.9E-01	2.5E-02
D4	3.45E+03	4.4E-02	1.2E+00	4.3E-02
D5	3.45E+03	2.0E-02	5.2E-01	2.3E-02
D6	3.45E+03	5.1E-02	1.7E+00	5.4E-02
E1	1.38E+04	2.2E-02	7.5E-01	2.5E-02
E2	1.38E+04	7.8E-03	3.4E-01	1.2E-02
E3	1.38E+04	7.5E-02	2.4E+00	7.5E-02

E4	1.38E+04	1.6E-02	4.7E-01	1.5E-02
E5	1.38E+04	3.3E-02	1.5E+00	4.0E-02
E6	1.38E+04	8.6E-03	2.6E-01	7.8E-03
F1	2.76E+04	7.2E-03	2.0E-01	7.4E-03
F2	2.76E+04	5.8E-03	1.9E-01	5.2E-03
F3	2.76E+04	4.2E-02	1.5E+00	4.4E-02
F4	2.76E+04	2.3E-02	8.7E-01	2.8E-02
F5	2.76E+04	3.8E-02	1.1E+00	3.7E-02
F6	2.76E+04	2.4E-02	7.3E-01	2.3E-02
G1	1.72E+03	3.9E-02	1.4E+00	4.2E-02
G2	1.72E+03	5.2E-02	1.3E+00	5.9E-02
G3	1.72E+03	5.6E-02	1.7E+00	5.2E-02
G4	1.72E+03	5.8E-02	2.0E+00	6.3E-02
G5	1.72E+03	5.8E-02	1.6E+00	6.0E-02
G6	1.72E+03	4.6E-02	2.2E+00	7.9E-02

Table A-2 Radish Interception Results continued.

A.1.2 Radish Absorption Results

Sample	Plant Count Rate (CPM)	Radish Count Rate (CPM)	Plant dry mass (g)	Radish dry mass (g)	Activity deposited (Bq)
A1	48902	1661	0.243	0.322	3.93E+03
A2	53301	2253	0.243	0.325	3.93E+03
A3	43378	1762	0.288	0.299	3.93E+03
A4	57239	1339	0.233	0.146	3.93E+03
A5	55346	1678	0.234	0.176	3.93E+03
A6	65176	4033	0.195	0.161	3.93E+03
B1	47927	1548	0.306	0.226	3.93E+03
B2	49527	1886	0.256	0.289	3.93E+03
B3	45558	10351	0.18	0.159	3.93E+03
B4	53958	2862	0.229	0.288	3.93E+03
B5	50860	4538	0.24	0.215	3.93E+03
B6	44453	1412	0.237	0.364	3.93E+03
C1	49140	1722	0.302	0.364	3.93E+03
C2	46459	1554	0.279	0.293	3.93E+03

C3	37294	1975	0.317	0.253	3.93E+03
C4	46191	1555	0.324	0.192	3.93E+03
C5	43970	2207	0.31	0.154	3.93E+03
C6	59651	3109	0.222	0.17	3.93E+03
D1	42942	2842	0.292	0.534	3.93E+03
D2	33954	1417	0.387	0.65	3.93E+03
D3	33422	2948	0.378	0.475	3.93E+03
D4	34202	2508	0.34	0.625	3.93E+03
D5	27453	1280	0.426	0.736	3.93E+03
D6	35370	3447	0.391	0.737	3.93E+03

Table A-3 Radish absorption results.

Sample Name	**Plant Activity, (Bq g^{-1})**	**Radish Activity, (Bq g^{-1})**	**ROR method**	**ROA method**	**IA method**
A1	1.27E+04	4.16E+02	96%	82%	96%
A2	1.38E+04	5.66E+02	94%	90%	94%
A3	1.12E+04	4.42E+02	93%	86%	94%
A4	1.48E+04	3.35E+02	94%	89%	95%
A5	1.43E+04	4.21E+02	90%	87%	91%
A6	1.69E+04	1.02E+03	96%	88%	96%
B1	1.24E+04	3.88E+02	98%	99%	98%
B2	1.28E+04	4.73E+02	98%	87%	98%
B3	1.18E+04	2.62E+03	96%	65%	97%
B4	1.40E+04	7.20E+02	98%	87%	98%
B5	1.32E+04	1.14E+03	97%	87%	98%
B6	1.15E+04	3.53E+02	96%	73%	97%
C1	1.27E+04	4.32E+02	97%	102%	97%
C2	1.20E+04	3.89E+02	98%	88%	98%
C3	9.65E+03	4.96E+02	98%	81%	99%
C4	1.20E+04	3.90E+02	97%	100%	97%
C5	1.14E+04	5.55E+02	99%	92%	99%
C6	1.54E+04	7.83E+02	97%	90%	97%
D1	1.11E+04	7.15E+02	97%	92%	97%
D2	8.78E+03	3.55E+02	97%	92%	97%
D3	8.65E+03	7.42E+02	97%	92%	98%
D4	8.85E+03	6.31E+02	95%	87%	95%

D5	7.10E+03	3.20E+02	98%	83%	98%
D6	9.15E+03	8.68E+02	97%	107%	96%

Table A-3 Radish absorption results continued.

A.1.3 Radish Translocation Results

Sample	Plant Count Rate (CPM)	Radish Count Rate (CPM)	Plant dry mass (g)	Radish dry mass (g)
A1	36613	2654	0.314	0.563
A2	29229	997	0.459	0.486
A3	32230	1086	0.415	0.54
A4	43134	1091	0.304	0.42
A5	29591	710	0.505	0.287
A6	30291	1028	0.366	0.661
B1	49949	4025	0.274	0.558
B2	36264	2863	0.332	0.727
B3	43004	923	0.327	0.364
B4	42482	3179	0.297	0.604
B5	38387	2754	0.344	0.614
B6	47630	3233	0.281	0.554
C1	43718	3865	0.292	0.657
C2	36317	3229	0.404	0.948
C3	35790	3355	0.385	0.845
C4	43444	2663	0.357	0.731
C5	31349	2770	0.425	0.735
C6	53596	4093	0.274	0.786

Table A-4 Radish translocation results.

Sample	Plant activity, (Bq g^{-1})	Radish activity, (Bq g^{-1})	Activity deposited (Bq)	ROR method	ROA method	CR method
A1	9.47E+03	6.68E+02	3.93E+03	11%	10%	7.05%
A2	7.56E+03	2.48E+02	3.93E+03	3%	3%	3.28%
A3	8.34E+03	2.71E+02	3.93E+03	4%	4%	3.25%
A4	1.12E+04	2.72E+02	3.93E+03	3%	3%	2.44%
A5	7.65E+03	1.76E+02	3.93E+03	1%	1%	2.29%
A6	7.84E+03	2.56E+02	3.93E+03	6%	4%	3.27%

B1	1.29E+04	1.01E+03	3.93E+03	14%	14%	7.85%
B2	9.38E+03	7.21E+02	3.93E+03	14%	13%	7.68%
B3	1.11E+04	2.30E+02	3.93E+03	2%	2%	2.06%
B4	1.10E+04	8.01E+02	3.93E+03	13%	12%	7.28%
B5	9.93E+03	6.93E+02	3.93E+03	11%	11%	6.98%
B6	1.23E+04	8.14E+02	3.93E+03	12%	11%	6.61%
C1	1.13E+04	9.74E+02	3.93E+03	16%	16%	8.61%
C2	9.39E+03	8.13E+02	3.93E+03	17%	20%	8.66%
C3	9.26E+03	8.45E+02	3.93E+03	17%	18%	9.13%
C4	1.12E+04	6.70E+02	3.93E+03	11%	12%	5.96%
C5	8.11E+03	6.97E+02	3.93E+03	13%	13%	8.60%
C6	1.39E+04	1.03E+03	3.93E+03	18%	21%	7.44%

Table A-4 Radish translocation results continued.

A.2 Beans

The averaged background CPM for each type of bean sample; bean pod, inedible plant tissue, and total plant were respectively 16, 12, 16.

A.2.1 Bean Interception Results

Sample	Count Rate (CPM)	Plant dry weight (g)	Plant Activity, (Bq g^{-1})	Leaf area, (cm^2)
A1	483	3.025	1.29E+02	93.23
A2	452	3.103	1.21E+02	92.47
A3	594	3.676	1.60E+02	110.11
A4	535	3.479	1.44E+02	106.51
A5	453	3.793	1.21E+02	110.33
A6	481	3.644	1.29E+02	110.61
B1	663	10.198	1.79E+02	272.77
B2	644	8.567	1.74E+02	243.22

B3	736	9.26	1.99E+02	260.66
B4	777	6.949	2.11E+02	193.43
B5	826	10.695	2.24E+02	287.59
B6	835	8.222	2.27E+02	253.2
C1	210	47.051	5.34E+01	393.83
C2	229	46.36	5.87E+01	338.17
C3	148	53.494	3.62E+01	353.02
C4	172	46.47	4.28E+01	288.5
C5	145	62.518	3.54E+01	437.96
C6	213	47.9	5.42E+01	340.46
D1	259	54.925	6.70E+01	325.73
D2	247	42.881	6.37E+01	305.76
D3	236	59.56	6.06E+01	359.88
D4	304	34.093	7.95E+01	216.81
D5	298	38.413	7.78E+01	291.53
D6	391	31.635	1.04E+02	297.12
E1	276	56.946	7.17E+01	384.59
E2	328	58.567	8.61E+01	400.59
E3	373	48.515	9.86E+01	351.47
E4	459	50.754	1.22E+02	362.11
E5	260	50.383	6.73E+01	354.32
E6	339	62.691	8.92E+01	446.1
F1	431	50.808	1.15E+02	331.72
F2	467	39.89	1.25E+02	258.02
F3	556	36.264	1.49E+02	318.94
F4	510	37.725	1.37E+02	326.82
F5	525	31.58	1.41E+02	196
F6	397	42.515	1.05E+02	270.82
G1	436	51.177	1.16E+02	367.15
G2	487	58.772	1.30E+02	345.31
G3	384	62.181	1.02E+02	397.95
G4	514	56.846	1.38E+02	402.07
G5	438	61.75	1.17E+02	426.98
G6	691	41.351	1.87E+02	354.88

Table A-5 Bean interception results.

Sample	Activity Deposited (Bq)	f_{LAI}	f_B	f_{LAI}
A1	3.92E+03	1.0E-01	5.6E-01	1.8E-01
A2	3.92E+03	9.5E-02	5.2E-01	1.7E-01
A3	3.92E+03	1.5E-01	6.9E-01	2.3E-01
A4	3.92E+03	1.3E-01	6.2E-01	2.0E-01
A5	3.92E+03	1.2E-01	5.2E-01	1.8E-01
A6	3.92E+03	1.2E-01	5.5E-01	1.8E-01
B1	1.06E+04	1.7E-01	2.8E-01	1.1E-01
B2	1.06E+04	1.4E-01	2.8E-01	9.7E-02
B3	1.06E+04	1.7E-01	3.2E-01	1.1E-01
B4	1.06E+04	1.4E-01	3.4E-01	1.2E-01
B5	1.06E+04	2.3E-01	3.6E-01	1.3E-01
B6	1.06E+04	1.8E-01	3.6E-01	1.2E-01
C1	6.03E+03	4.2E-01	1.5E-01	1.8E-01
C2	6.03E+03	4.5E-01	1.6E-01	2.3E-01
C3	6.03E+03	3.2E-01	1.0E-01	1.5E-01
C4	6.03E+03	3.3E-01	1.2E-01	1.9E-01
C5	6.03E+03	3.7E-01	9.9E-02	1.4E-01
C6	6.03E+03	4.3E-01	1.5E-01	2.1E-01
D1	1.21E+04	3.1E-01	9.4E-02	1.6E-01
D2	1.21E+04	2.3E-01	8.9E-02	1.3E-01
D3	1.21E+04	3.0E-01	8.5E-02	1.4E-01
D4	1.21E+04	2.2E-01	1.1E-01	1.8E-01
D5	1.21E+04	2.5E-01	1.1E-01	1.4E-01
D6	1.21E+04	2.7E-01	1.5E-01	1.5E-01
E1	2.41E+04	1.7E-01	5.0E-02	7.4E-02
E2	2.41E+04	2.1E-01	6.0E-02	8.8E-02
E3	2.41E+04	2.0E-01	6.9E-02	9.5E-02
E4	2.41E+04	2.6E-01	8.6E-02	1.2E-01
E5	2.41E+04	1.4E-01	4.7E-02	6.7E-02
E6	2.41E+04	2.3E-01	6.3E-02	8.8E-02
F1	4.82E+04	1.2E-01	4.0E-02	6.2E-02
F2	4.82E+04	1.0E-01	4.4E-02	6.8E-02
F3	4.82E+04	1.1E-01	5.2E-02	6.0E-02
F4	4.82E+04	1.1E-01	4.8E-02	5.5E-02

F5	4.82E+04	9.2E-02	4.9E-02	7.9E-02
F6	4.82E+04	9.3E-02	3.7E-02	5.8E-02
G1	9.64E+04	6.2E-02	2.0E-02	2.8E-02
G2	9.64E+04	7.9E-02	2.3E-02	3.9E-02
G3	9.64E+04	6.6E-02	1.8E-02	2.8E-02
G4	9.64E+04	8.1E-02	2.4E-02	3.4E-02
G5	9.64E+04	7.5E-02	2.0E-02	3.0E-02
G6	9.64E+04	8.0E-02	3.3E-02	3.8E-02

Table A-5 Bean interception results continued.

A.2.2 Bean Absorption Results

Sample	Plant Count Rate (CPM)	Bean pod Count Rate (CPM)	Plant dry mass (g)	Bean pod dry mass (g)	Activity deposited (Bq)
A1	3675	119	9.248	8.075	1.13E+04
A2	2902	154	11.128	11.75	1.13E+04
A3	3272	132	11.703	10.428	1.13E+04
A4	3309	153	10.665	6.941	1.13E+04
A5	3942	271	11.06	7.35	1.13E+04
A6	1566	186	14.978	6.097	1.13E+04
B1	4800	235	7.779	9.024	1.13E+04
B2	2747	154	14.235	11.727	1.13E+04
B3	2810	123	12.55	12.756	1.13E+04
B4	2027	77	15.718	14.049	1.13E+04
B5	3387	74	15.092	14.927	1.13E+04
B6	3286	226	10.75	9.305	1.13E+04
C1	2604	129	12.961	30.486	1.13E+04
C2	1992	117	12.624	39.818	1.13E+04
C3	3809	189	10.824	22.776	1.13E+04
C4	2145	195	12.596	32.284	1.13E+04
C5	1585	105	18.089	38.074	1.13E+04
C6	2730	216	10.299	26.415	1.13E+04
D1	3675	119	9.248	8.075	1.13E+04
D2	2902	154	11.128	11.75	1.13E+04
D3	3272	132	11.703	10.428	1.13E+04

D4	3309	153	10.665	6.941	1.13E+04
D5	3942	271	11.06	7.35	1.13E+04
D6	1566	186	14.978	6.097	1.13E+04

Table A-6 Bean absorption results.

Sample	Plant Activity, (Bq g^{-1})	Bean pod Activity, (Bq g^{-1})	ROR method	ROA method	IA method
A1	1.0E+03	2.7E+01	98%	86%	98%
A2	8.1E+02	3.6E+01	96%	83%	97%
A3	9.1E+02	3.0E+01	99%	97%	99%
A4	9.2E+02	3.6E+01	98%	89%	99%
A5	1.1E+03	6.6E+01	96%	112%	96%
A6	4.3E+02	4.4E+01	96%	60%	97%
B1	1.3E+03	5.6E+01	99%	96%	99%
B2	7.7E+02	3.6E+01	98%	100%	98%
B3	7.8E+02	2.8E+01	98%	90%	98%
B4	5.6E+02	1.6E+01	98%	80%	99%
B5	9.5E+02	1.6E+01	99%	128%	98%
B6	9.2E+02	5.4E+01	98%	91%	98%
C1	7.3E+02	3.0E+01	93%	91%	93%
C2	5.5E+02	2.7E+01	91%	71%	93%
C3	1.1E+03	4.5E+01	98%	110%	97%
C4	6.0E+02	4.6E+01	92%	79%	93%
C5	4.4E+02	2.4E+01	87%	78%	89%
C6	7.6E+02	5.2E+01	96%	81%	96%
D1	1.0E+03	2.7E+01	98%	86%	98%
D2	8.1E+02	3.6E+01	96%	83%	97%
D3	9.1E+02	3.0E+01	99%	97%	99%
D4	9.2E+02	3.6E+01	98%	89%	99%
D5	1.1E+03	6.6E+01	96%	112%	96%
D6	4.3E+02	4.4E+01	96%	60%	97%

Table A-6 Bean absorption results continued.

A.2.3 Bean Translocation Results

Sample	Plant count rate (CPM)	Bean pod count rate (CPM)	Plant dry mass (g)	Bean pod dry mass (g)
A1	1162	73	10.764	26.513
A2	1185	53	14.054	30.387
A3	1046	73	12.662	25.918
A4	1231	62	9.408	19.684
A5	1116	90	13.841	26.508
A6	1334	65	9.495	19.888
B1	3357	403	11.202	23.885
B2	1479	181	21.757	37.664
B3	1645	188	19.726	35.753
B4	1923	213	17.754	35.354
B5	2041	143	14.824	33.326
B6	2763	240	15.017	35.86
C1	2203	88	16.889	41.881
C2	2592	170	12.163	35.777
C3	1507	109	16.036	36.968
C4	2398	112	15.019	36.026
C5	4516	155	8.541	26.737
C6	2207	160	13.049	30.345
D1	3612	133	12.045	13.929
D2	2904	65	11.376	27.875
D3	1875	38	16.217	38.358
D4	2483	61	14.101	20.818
D5	2228	52	15.711	36.302
D6	3575	69	13.979	34.018
E1	1872	23	20.897	33.747
E2	3537	25	11.42	26.551
E3	2508	22	16.094	28.97
E4	3270	21	9.686	22.551
E5	2415	27	18.376	23.65
E6	1684	21	23.359	33.436

Table A-7 Bean translocation results.

Sample	Plant activity, (Bq g^{-1})	Bean pod activity, (Bq g^{-1})	Activity deposited, (Bq)	ROR method	ROA method	CR method
A1	3.2E+02	1.5E+01	4.35E+03	11%	9%	4.8%

A2	3.3E+02	1.0E+01	4.35E+03	6%	7%	3.2%
A3	2.9E+02	1.5E+01	4.35E+03	10%	9%	5.3%
A4	3.4E+02	1.3E+01	4.35E+03	7%	6%	3.7%
A5	3.1E+02	2.0E+01	4.35E+03	11%	12%	6.4%
A6	3.7E+02	1.3E+01	4.35E+03	7%	6%	3.6%
B1	9.4E+02	9.9E+01	1.12E+04	18%	21%	10.6%
B2	4.1E+02	4.3E+01	1.12E+04	15%	14%	10.4%
B3	4.6E+02	4.5E+01	1.12E+04	15%	14%	9.8%
B4	5.3E+02	5.1E+01	1.12E+04	16%	16%	9.5%
B5	5.7E+02	3.3E+01	1.12E+04	12%	10%	5.8%
B6	7.7E+02	5.8E+01	1.12E+04	15%	18%	7.5%
C1	6.1E+02	1.9E+01	1.13E+04	7%	7%	3.1%
C2	7.2E+02	4.0E+01	1.13E+04	14%	13%	5.5%
C3	4.2E+02	2.5E+01	1.13E+04	12%	8%	5.9%
C4	6.7E+02	2.5E+01	1.13E+04	8%	8%	3.8%
C5	1.3E+03	3.6E+01	1.13E+04	8%	9%	2.9%
C6	6.1E+02	3.7E+01	1.13E+04	12%	10%	6.1%
D1	1.0E+03	3.1E+01	1.15E+04	3%	4%	3.0%
D2	8.1E+02	1.3E+01	1.15E+04	4%	3%	1.7%
D3	5.2E+02	6.6E+00	1.15E+04	3%	2%	1.3%
D4	6.9E+02	1.2E+01	1.15E+04	3%	2%	1.8%
D5	6.2E+02	1.0E+01	1.15E+04	4%	3%	1.6%
D6	1.0E+03	1.4E+01	1.15E+04	3%	4%	1.4%
E1	5.2E+02	2.8E+00	1.16E+04	1%	1%	0.5%
E2	9.9E+02	3.3E+00	1.16E+04	1%	1%	0.3%
E3	7.0E+02	2.5E+00	1.16E+04	1%	1%	0.4%
E4	9.1E+02	2.3E+00	1.16E+04	1%	0%	0.2%
E5	6.7E+02	3.8E+00	1.16E+04	1%	1%	0.6%
E6	4.7E+02	2.3E+00	1.16E+04	1%	1%	0.5%

Table A-7 Bean translocation results continued.

A.3 Wheat

The averaged background CPM for each type of wheat sample; grain, inedible plant tissue, and total plant were respectively 37.5, 12.5, 13.5.

A.3.1 Wheat Interception Results

Sample	Count Rate (CPM)	Plant weight (dry), g	Plant activity, (Bq g^{-1})	Leaf area, (cm^2)
A1	202	9.756	6.2E+01	102.24
A2	221	7.014	6.8E+01	100.29
A3	187	7.366	5.7E+01	97.98
A4	154	7.188	4.6E+01	119.71
A5	220	8.267	6.8E+01	107.62
A6	217	7.601	6.7E+01	106.8
B1	347	11.756	1.1E+02	104.57
B2	397	14.745	1.3E+02	91.51
B3	406	14.212	1.3E+02	101.89
B4	759	12.854	2.4E+02	113.67
B5	456	14.065	1.5E+02	97.61
B6	323	12.22	1.0E+02	82.68
C1	261	29.918	8.1E+01	116.85
C2	273	23.925	8.5E+01	128.25
C3	239	28.626	7.4E+01	99.22
C4	226	34.113	7.0E+01	114.36
C5	213	28.224	6.6E+01	114.1
C6	244	21.522	7.6E+01	163.45
D1	401	28.016	1.3E+02	106.77
D2	267	43.613	8.3E+01	102.76
D3	359	28.915	1.1E+02	80.16
D4	451	31.073	1.4E+02	103.54
D5	303	41.388	9.5E+01	89.71
D6	328	42.367	1.0E+02	98.42
E1	574	25.056	1.8E+02	91.82
E2	529	28.898	1.7E+02	107.05
E3	451	27.689	1.4E+02	89.53
E4	610	32.196	2.0E+02	130.14
E5	622	23.602	2.0E+02	108.06
E6	517	27.367	1.7E+02	101.47
F1	858	22.113	2.8E+02	85.77

F2	847	26.135	2.7E+02	109.33
F3	660	24.213	2.1E+02	87.82
F4	795	31.606	2.6E+02	105.59
F5	840	29.391	2.7E+02	109.46
F6	864	23.577	2.8E+02	122.78
G1	1088	23.155	3.5E+02	102.24
G2	1199	23.093	3.9E+02	100.29
G3	996	26.997	3.2E+02	97.98
G4	1028	29.098	3.3E+02	119.71
G5	1365	31.234	4.4E+02	107.62
G6	1209	31.432	3.9E+02	106.8

Table A-8 Wheat interception results.

Sample	**Activity Applied (Bq)**	**f_{LAI}**	**F_M**	**f_{LAI}**
A1	3.92E+03	1.5E-01	2.7E-01	no data
A2	3.92E+03	1.2E-01	2.9E-01	no data
A3	3.92E+03	1.1E-01	2.5E-01	no data
A4	3.92E+03	8.5E-02	2.0E-01	no data
A5	3.92E+03	1.4E-01	2.9E-01	no data
A6	3.92E+03	1.3E-01	2.9E-01	no data
B1	1.06E+04	1.2E-01	1.7E-01	2.0E-01
B2	1.06E+04	1.7E-01	2.0E-01	2.9E-01
B3	1.06E+04	1.7E-01	2.1E-01	3.0E-01
B4	1.06E+04	3.0E-01	3.9E-01	4.2E-01
B5	1.06E+04	1.9E-01	2.3E-01	3.0E-01
B6	1.06E+04	1.2E-01	1.6E-01	1.9E-01
C1	4.06E+03	6.0E-01	3.4E-01	9.7E-01
C2	4.06E+03	5.0E-01	3.5E-01	9.3E-01
C3	4.06E+03	5.2E-01	3.1E-01	8.7E-01
C4	4.06E+03	5.9E-01	2.9E-01	8.7E-01
C5	4.06E+03	4.6E-01	2.7E-01	7.9E-01
C6	4.06E+03	4.0E-01	3.1E-01	8.2E-01
D1	8.13E+03	4.4E-01	2.6E-01	6.3E-01
D2	8.13E+03	4.5E-01	1.7E-01	5.9E-01
D3	8.13E+03	4.0E-01	2.4E-01	6.9E-01
D4	8.13E+03	5.5E-01	3.0E-01	8.1E-01

D5	8.13E+03	4.8E-01	2.0E-01	7.2E-01
D6	8.13E+03	5.4E-01	2.1E-01	5.6E-01
E1	1.63E+04	2.8E-01	1.9E-01	4.5E-01
E2	1.63E+04	3.0E-01	1.8E-01	5.0E-01
E3	1.63E+04	2.4E-01	1.5E-01	5.2E-01
E4	1.63E+04	3.9E-01	2.0E-01	6.3E-01
E5	1.63E+04	2.9E-01	2.1E-01	5.5E-01
E6	1.63E+04	2.8E-01	1.7E-01	4.8E-01
F1	3.25E+04	1.9E-01	1.4E-01	3.5E-01
F2	3.25E+04	2.2E-01	1.4E-01	3.5E-01
F3	3.25E+04	1.6E-01	1.1E-01	3.0E-01
F4	3.25E+04	2.5E-01	1.3E-01	3.2E-01
F5	3.25E+04	2.5E-01	1.4E-01	3.8E-01
F6	3.25E+04	2.0E-01	1.5E-01	3.4E-01
G1	6.45E+04	1.3E-01	9.2E-02	2.5E-01
G2	6.45E+04	1.4E-01	1.0E-01	2.2E-01
G3	6.45E+04	1.4E-01	8.5E-02	2.6E-01
G4	6.45E+04	1.5E-01	8.7E-02	2.4E-01
G5	6.45E+04	2.1E-01	1.2E-01	3.3E-01
G6	6.45E+04	1.9E-01	1.0E-01	2.6E-01

Table A-8 Wheat interception results continued.

A.3.2 Wheat Absorption Results

Sample	Plant count rate (CPM)	Grain count rate (CPM)	Plant dry mass (g)	Grain dry mass (g)	Activity Deposited (Bq)
A1	954	92	14.256	7.325	4.54E+03
A2	924	42	16.212	10.895	4.54E+03
A3	1251	73	13.667	7.827	4.54E+03
A4	1025	129	12.914	8.698	4.54E+03
A5	919	70	17.704	10.013	4.54E+03
A6	1195	90	12.929	7.97	4.54E+03
B1	1003	75	13.839	8.433	4.54E+03
B2	1258	172	12.291	8.246	4.54E+03
B3	1308	105	11.873	9.777	4.54E+03
B4	1273	102	11.775	7.571	4.54E+03

B5	1115	82	11.898	9.717	4.54E+03
B6	1002	127	14.242	9.943	4.54E+03
C1	1089	181	13.747	24.302	4.54E+03
C2	845	184	14.43	29.128	4.54E+03
C3	911	138	14.239	27.378	4.54E+03
C4	796	219	13.391	22.168	4.54E+03
C5	855	162	12.894	23.648	4.54E+03
C6	824	178	12.806	23.451	4.54E+03

Table A-9 Wheat absorption results.

Sample	Plant activity, (Bq g^{-1})	Grain activity, (Bq g^{-1})	ROR method	ROA method	IA method
A1	3.1E+02	2.4E+01	97%	103%	97%
A2	3.0E+02	8.9E+00	98%	111%	97%
A3	4.2E+02	1.8E+01	99%	129%	98%
A4	3.4E+02	3.5E+01	96%	103%	96%
A5	3.0E+02	1.7E+01	98%	122%	97%
A6	4.0E+02	2.3E+01	97%	117%	96%
B1	3.3E+02	1.9E+01	99%	105%	99%
B2	4.2E+02	4.8E+01	98%	122%	97%
B3	4.4E+02	2.8E+01	99%	120%	99%
B4	4.2E+02	2.7E+01	99%	114%	99%
B5	3.7E+02	2.1E+01	98%	101%	98%
B6	3.3E+02	3.5E+01	98%	111%	98%
C1	3.6E+02	5.1E+01	97%	137%	96%
C2	2.8E+02	5.2E+01	92%	121%	90%
C3	3.0E+02	3.8E+01	95%	117%	94%
C4	2.6E+02	6.2E+01	94%	107%	93%
C5	2.8E+02	4.5E+01	96%	103%	96%
C6	2.7E+02	5.0E+01	90%	102%	88%

Table A-9 Wheat absorption results continued.

A.3.3 Wheat Translocation Results

Sample	Plant count rate (CPM)	Grain count rate (CPM)	Plant dry mass (g)	Grain dry mass (g)

TLOC A1	806	309	13.552	17.462
TLOC A2	794	297	11.957	16.176
TLOC A3	806	356	11.728	15.133
TLOC A4	841	122	14.714	26.644
TLOC A5	655	310	16.135	23.415
TLOC A6	754	228	12.785	19.838
TLOC B1	719	357	16.084	19.123
TLOC B2	865	256	14.556	36.717
TLOC B3	1081	351	12.454	17.294
TLOC B4	790	262	13.377	24.424
TLOC B5	804	300	13.295	20.286
TLOC B6	917	158	12.821	22.575
TLOC C1	821	139	15.56	24.325
TLOC C2	606	148	19.539	35.962
TLOC C3	961	125	14.059	26.8
TLOC C4	828	114	15.147	27.169
TLOC C5	702	121	18.253	30.488
TLOC C6	789	228	14.258	22.828
TLOC D1	811	145	14.631	22.384
TLOC D2	1269	179	11.474	15.808
TLOC D3	1189	148	9.926	17.289
TLOC D4	764	200	14.65	21.415
TLOC D5	783	116	15.848	28.722
TLOC D6	752	177	14.973	21.123
TLOC E1	1249	77	12.45	19.017
TLOC E2	1120	121	14.299	19.382
TLOC E3	1058	111	13.51	20.071
TLOC E4	1301	48	11.212	18.294
TLOC E5	1255	113	11.926	16.897
TLOC E6	937	123	14.108	19.644

Table A-10 Wheat translocation results.

Sample	Plant activity, (Bq g^{-1})	Grain activity, (Bq g^{-1})	Activity deposited, (Bq)	ROR method	ROA method	CR method
TLOC A1	2.6E+02	9.0E+01	4.50E+03	30%	35%	34.0%
TLOC A2	2.6E+02	8.6E+01	4.50E+03	31%	31%	33.2%
TLOC A3	2.6E+02	1.0E+02	4.50E+03	34%	35%	39.4%

TLOC A4	2.8E+02	3.3E+01	4.50E+03	18%	20%	12.0%
TLOC A5	2.1E+02	9.0E+01	4.50E+03	38%	47%	42.5%
TLOC A6	2.5E+02	6.5E+01	4.50E+03	29%	29%	26.5%
TLOC B1	2.3E+02	1.0E+02	4.58E+03	35%	44%	44.6%
TLOC B2	2.8E+02	7.4E+01	4.58E+03	40%	59%	25.9%
TLOC B3	3.6E+02	1.0E+02	4.58E+03	28%	39%	28.6%
TLOC B4	2.6E+02	7.5E+01	4.58E+03	35%	40%	29.2%
TLOC B5	2.6E+02	8.7E+01	4.58E+03	34%	39%	33.1%
TLOC B6	3.0E+02	4.4E+01	4.58E+03	20%	22%	14.6%
TLOC C1	2.7E+02	3.8E+01	4.58E+03	18%	20%	14.2%
TLOC C2	2.0E+02	4.1E+01	4.58E+03	28%	32%	21.0%
TLOC C3	3.2E+02	3.4E+01	4.58E+03	17%	20%	10.7%
TLOC C4	2.7E+02	3.1E+01	4.58E+03	17%	18%	11.3%
TLOC C5	2.3E+02	3.3E+01	4.58E+03	19%	22%	14.4%
TLOC C6	2.6E+02	6.5E+01	4.58E+03	29%	32%	25.3%
TLOC D1	2.7E+02	4.0E+01	4.64E+03	19%	19%	15.1%
TLOC D2	4.2E+02	5.0E+01	4.64E+03	14%	17%	11.9%
TLOC D3	4.0E+02	4.1E+01	4.64E+03	15%	15%	10.4%
TLOC D4	2.5E+02	5.7E+01	4.64E+03	25%	26%	22.8%
TLOC D5	2.6E+02	3.1E+01	4.64E+03	18%	19%	12.2%
TLOC D6	2.5E+02	5.0E+01	4.64E+03	22%	23%	20.3%
TLOC E1	4.2E+02	2.0E+01	4.64E+03	7%	8%	4.7%
TLOC E2	3.7E+02	3.3E+01	4.64E+03	11%	14%	8.8%
TLOC E3	3.5E+02	3.0E+01	4.64E+03	11%	13%	8.5%
TLOC E4	4.3E+02	1.1E+01	4.64E+03	4%	4%	2.5%
TLOC E5	4.2E+02	3.0E+01	4.64E+03	9%	11%	7.3%
TLOC E6	3.1E+02	3.3E+01	4.64E+03	13%	14%	10.8%

Table A-19 Wheat translocation results continued.