Modelling Plant Uptake of Sulphur-35

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ABSTRACT

Sulphur-35 is discharged as a gas from UK Magnox and AGR nuclear reactors in two important forms. The first, $CO^{35}S$ (carbonyl sulphide), is released routinely during reactor operation while the second, $H_2{}^{35}S$ (hydrogen sulphide), is released primarily from Magnox reactors when reactor maintenance is carried out. The uptake of gaseous forms of ^{35}S (sulphur-35) by plant leaves is known to be an important mechanism leading to the contamination of crops. It is thought that this route of exposure may not be adequately modelled in the HPA-RPD foodchain model FARMLAND and therefore a new model SGAS has been developed. This report describes SGAS and the data on which it has been based. It also highlights some of the problems associated with using these data and identifies possible areas of future work.

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Approval: March 2006
Publication: May 2006
£14.00
ISBN 0 85951 576 1

EXECUTIVE SUMMARY

Sulphur-35 is discharged as a gas from UK Magnox and AGR nuclear reactors in two important forms. The first, $CO^{35}S$, is released routinely during reactor operation while the second, $H_2{}^{35}S$, is released primarily from Magnox reactors when reactor maintenance is carried out.

For some time it has been understood that the uptake of gaseous forms of ^{35}S via plant leaves is a potentially important contamination pathway. This uptake route may not be modelled adequately in the HPA-RPD's* terrestrial foodchain model FARMLAND which assumes that ^{35}S is released in particulate form. This report identifies some of the more important sources of data related to the uptake and distribution of sulphur within plants. Using an empirical approach, the report goes on to describe new models that better represent the limited experimental data. The first represents plants for which direct contamination of the edible parts occurs and the second takes account of the process of translocation where activity is moved from inedible to edible parts. The revised models, collectively referred to as SGAS, have been applied to a number of release scenarios and give rise to activity concentrations in foods that are generally greater than those predicted using the existing FARMLAND models. For continuos releases to atmosphere SGAS predicts activity concentrations that are less than a factor of two higher than those predicted by FARMLAND. For short duration releases the differences are greater, with activity concentrations in leaves from SGAS being greater than FARMLAND by a factor of about 10 at 60 days after the release. For the spreading of sewage sludge and application of contaminated irrigation water, the results are more similar.

Given the many uncertainties associated with the SGAS models described in this report it is recommended that they are only used as screening tools, with particular application for short duration planned releases.

There are a number of areas that would benefit from further experimental study. In particular, the influence of realistic background air concentrations of $CO^{35}S$ and other forms of sulphur on the uptake mechanisms of plants could be investigated. Such studies should include a sample size that is large enough to overcome any inherent variability in the measurements to ensure that trends in the uptake process can be identified.

* HPA-RPD is the Radiation Protection Division of the Health Protection Agency, formerly the National Radiological Protection Board.

CONTENTS

1 INTRODUCTION

Sulphur-35 is discharged as a gas from UK Magnox and Advanced Gas-Cooled (AGR) nuclear reactors in two important forms (Bailey, 1975). The first, $CO^{35}S$, is released routinely during reactor operation while the second, $H_2{}^{35}S$, is released primarily from Magnox reactors when reactor maintenance is carried out.

For some time it has been understood that the uptake of gaseous forms of ^{35}S via plant leaves is a potentially important contamination pathway. This uptake route may not be modelled adequately in the HPA-RPD’s terrestrial foodchain model FARMLAND which assumes that ^{35}S is released in particulate form. Despite this FARMLAND model predictions for continuous releases compare reasonably well with experimental data. However, this is not the case for short duration releases. Therefore, the aim of this project was to produce a model that could better predict the uptake of ^{35}S by plants and that was applicable to both short and continuous releases. The report identifies some of the more important sources of data related to the uptake and distribution of sulphur within plants and considers the use of the SULPHUR model developed by Nuclear Electric plc (Pearce, 1991). This model was developed using data from a set of laboratory experiments where plants were exposed to short duration releases. There was considerable uncertainty in these data and SULPHUR conservatively assumed that the only loss following uptake was through radioactive decay. Because of this assumption the SULPHUR model could not be applied to continuous release scenarios because it significantly over estimated activity concentrations when compared to the data from field experiments. An underlying problem is that the plant probably regulates the uptake of sulphur. Therefore, in an environment that is initially sulphur free (ie the laboratory) the plant will take up discharged sulphur rapidly and retain it. However, in an environment with an ambient level of sulphur (eg around a nuclear site) uptake is regulated by the plant ie levels in the plant do not continuously rise.

Using an empirical approach, the report describes new models, collectively referred to as SGAS, that might be implemented to give reasonable predictions of the sulphur activity in plants following short and continuous releases. However, it is important to note that the experimental data on which the models are based are limited, include considerable uncertainties and in some cases appear to conflict. Consequently, this has a direct impact on the reliability of the models and it is recommended that they are only used for screening studies. The implications of the suggested modelling changes for dose assessments are discussed and areas of future work identified.

2 EXPERIMENTAL DATA

This section describes the two principal sources of data used in the development of new models. Although additional data sources have been used to guide the choice of transfer rates for specific processes (Section 5.2), ultimately values for these rates have been chosen to give model predictions that are a reasonable fit to the data described in this section.

2.1 Activity concentrations in crops following continuous discharges

Field studies have been carried out to investigate the uptake of $CO^{35}S$ by plants in the vicinity of Hinkley Point power station (Kluczewski, 1986). From this study estimates were made of the activity concentration in edible parts of the plant per unit air concentration for green vegetables and root vegetables, see Table 1. The green vegetable category includes broad beans as well as leafy green vegetables. Values for broad beans were 99 and 213 Bq kg^{-1} per Bq m^{-3}. In this study measurements of ambient levels of ^{35}S due to routine discharges were of the order of 1 10^{2} mBq m^{-3}.

Table 1 Activity concentrations of ^{35}S in plants per unit air concentration

		Bq kg^{-1} fresh weight plant per Bq m^{-3} of air		
	Number of samples	Geometric mean	Extreme value (99th percentile)	Geometric standard deviation
Green vegetables	6	150	280	1.2
Root vegetables	6	58	190	1.5

2.2 Partitioning of activity in crops following short duration releases

Information on the uptake of $CO^{35}S$ and $H_2{}^{35}S$ is also available for short duration releases (Collins, 1996A; Collins, 1996B). Studies were carried out within the laboratory to investigate uptake and retention within cabbage, beans, potatoes and wheat following fumigation at various times before harvest. Measurements show the partitioning of activity within plants at various times (1 day to several months) after fumigation for plants at various stages of development. However, no convincing evidence was found to suggest that the loss of ^{35}S from plants occurs by any other mechanism than radioactive decay. An attempt has been made in this report to summarise the partitioning results, see Tables 2a to 2d.

Table 2a Partitioning of $CO^{35}S$ by plants one day after fumigation

Plant	Percentage of total activity of $CO^{35}S$ in plant				
Cabbage	Leaf	Root	Stem		
	70-90	66-75 of rest	25-34 of rest		
Wheat	Leaf	Root	Stem	Ear	
	2-70	50 of rest	50 of rest	0-40	
Bean	Leaf	Root	Stem	Bean pod	Seed
	45-80	33 of rest	33 of rest	33 of rest	0-10
Potato	Leaf	Root	Stem	New tuber	Old tuber
	On average about 50% of the initial uptake is recovered in the tubers				

Table 2b Partitioning of $CO^{35}S$ by plants 3 weeks to 4 months after fumigation

Plant	Percentage of total activity of $CO^{35}S$ in plant				
Cabbage	Leaf	Root	Stem		
	40-80	50 of rest	50 of rest		
Wheat	Leaf	Root	Stem	Ear	
	10 of rest	45 of rest	45 of rest	20-60	
Bean	Leaf	Root	Stem	Bean pod	Seed
	2-35	50 of rest	25 of rest	25 of rest	20-40
Potato	Leaf	Root	Stem	New tuber	Old tuber
	On average about 50% of the initial uptake is recovered in the tubers				

Table 2c Partitioning of $H_2{}^{35}S$ by plants one day after fumigation

Plant	Percentage of total activity of $H_2{}^{35}S$ in plant				
Cabbage	Leaf	Root	Stem		
	75-90	50 of rest	50 of rest		
Wheat	Leaf	Root	Stem	Ear	
	10-75	50 of rest	50 of rest	0-45	
Bean	Leaf	Root	Stem	Bean pod	Seed
	50-75	33 of rest	33 of rest	33 of rest	0-10
Potato	Leaf	Root	Stem	New tuber	Old tuber
	On average about 50% of the initial uptake is recovered in the tubers				

Table 2d Partitioning of $H_2{}^{35}S$ by plants 3 weeks to 4 months after fumigation

Plant	Percentage of total activity of $H_2{}^{35}S$ in plant				
Cabbage	Leaf	Root	Stem		
	50-80	50 of rest	50 of rest		
Wheat	Leaf	Root	Stem	Ear	
	20 of rest	40 of rest	40 of rest	20-55	
Bean	Leaf	Root	Stem	Bean pod	Seed
	5-20	50 of rest	25 of rest	25 of rest	25-40
Potato	Leaf	Root	Stem	New tuber	Old tuber
	On average about 50% of the initial uptake is recovered in the tubers				

In addition Pearce (1994) used the data from Collins (1996A; 1996B) to calculate the possible range of activity concentrations in the edible crop at harvest following exposure at various times before harvest. Although there is considerable variation in these activity concentrations they can be used to give an indication of the upper level of activity that might be measured in each crop following exposure. For $CO^{35}S$ these levels are of the order of 1 Bq kg^{-1} per Bq m^{-2} for cabbage, 0.5 Bq kg^{-1} per Bq m^{-2} for bean, 1 Bq kg^{-1} per Bq m^{-2} for wheat and 0.2 Bq kg^{-1} per Bq m^{-2} for potatoes. From the data it is difficult to draw any conclusions about when these upper levels are likely to occur in terms of days before harvest. However, plant activity and growth, screening of edible plant parts and the mass of edible parts are all contributing factors.

2.3 Interpreting the data

Previous studies of the uptake and transfer of ^{35}S by plants have demonstrated that it is a complex process that depends on plant physiology and various environmental conditions. It is very difficult to control all these parameters even under laboratory conditions and hence significant variability is seen in experimental results. This is also true of the studies referenced in Sections 2.1 and 2.2. Nevertheless, these studies have been used to identify general trends in the uptake and transfer processes that could be represented by a model.

In developing a revised model the data from Collins (1996A; 1996B) were used to model the relative activity concentrations in various parts of the plant and the data from Kluczewski (1986) to set a limit on the build up of activity in plants continuously exposed over longer periods of time. Data for the partitioning of $H_2{}^{35}S$ following a short release appear to follow similar patterns to that for $CO^{35}S$. There are no data available for the build up of $H_2{}^{35}S$ in plants following a continuous release but releases of this form of sulphur are only known to occur over short time periods.

Initial comparisons suggest there is a conflict between the two sets of data. If the only loss of ^{35}S from the plant occurs by radioactive decay as suggested by the data from Collins (1996A; 1996B) then there should be a build up of activity in plants that are continually exposed. This will result in activity concentrations that are much greater than those derived from field studies where crops are exposed to continuous releases.

However, other researchers have measured sulphur-containing gases emanating from plants. In Coughtry (1983) losses from plant leaves of the order of 15% to 25% are quoted. It has been suggested (Stewart, 1999) that these losses might occur from labile pools of sulphur. Within these pools sulphur becomes fixed over time and hence losses to the atmosphere gradually reduce. These apparent discrepancies probably arise as a consequence of the plant's ability to control the uptake process depending on its requirements.

3 FARMLAND

The FARMLAND model (Brown, 1995) can be used to predict the activity concentration in plants following the deposition of ^{35}S released to the atmosphere. However, the model assumes that ^{35}S is released in particulate form. The majority of these particles are deposited on the soil while material deposited on the plant is subject to weathering processes as well as uptake. As a consequence of weathering a significant amount of sulphur is lost from the plant to the soil which always contains most of the deposited sulphur. However, for gaseous forms of sulphur experimental data suggest that uptake occurs directly into the plant leaves rather than following deposition onto the plant surface. As a result less sulphur is subject to the weathering process. In fact the most likely loss mechanism for gaseous forms of sulphur from the plant is back to the atmosphere via the leaves. Experimental data also indicate that deposition to soil is much less significant for $CO^{35}S$ and $H_2{}^{35}S$ and hence the activity in the soil is expected to be much lower than that predicted by FARMLAND.

To estimate the activity concentration per unit air concentration using FARMLAND a deposition velocity must be used. This parameter determines the fraction of the activity concentration in air that is deposited on the ground in unit time. An interception factor is then used to estimate how much of this material is deposited on the plant and how much on the soil. The value of the deposition velocity for particles of ^{35}S typically used in assessments is 1 10^{-3} m s^{-1}. This leads to predictions of activity concentrations in the edible plant at harvest (assuming a growing period of 120 days) of 4.5 10^{2} Bq kg^{-1} per Bq m^{-3} for green vegetables and 9.8 10^{1} Bq kg^{-1} per Bq m^{-3} for root vegetables following a continuous release to atmosphere. Although the FARMLAND results compare reasonably well with the values presented in Table 1, the modelling approach adopted would suggest that this is somewhat by chance and that some cancellation of errors has occurred in their derivation.

If the default FARMLAND model is run for a short duration release the predictions of activity concentration in the plant are less representative of measured values. FARMLAND predicts lower activities in green vegetables and root vegetables and a partitioning of these activities that dose not represent the findings of experiments conducted by Collins (1996A; 1996B). If FARMLAND is adjusted to fit the short duration release data it is no longer capable of predicting the activity concentration in the plant following a continuous release. In this case activity concentrations are greatly overestimated.

4 NUCLEAR ELECTRIC MODEL - SULPHUR

The most recent version of the model developed by Nuclear Electric (Pearce, 1994) is based closely on the studies carried out by Imperial College (Collins, 1996A; Collins, 1996B). Model parameters were adjusted so that model predictions more closely represent the partitioning of activity measured in the experiments. This included setting losses from the plant to zero such that radioactive decay was the only process by which activity in the plant decreased. The model runs are based on a spike release of ^{35}S. However, when the model is used to predict the activity in plants following a continuous release they overestimate the measured values (Kluczewski, 1986) considerably because so little activity is being lost from the plant.

5 MODELLING ^{35}S UPTAKE BY CROPS

For the models used in dose assessments it is only necessary to predict the general trend in the movement of radionuclides through the environment. The aim is to estimate the activity concentration when the food is consumed and not necessarily all of the processes involved in the transfer. It is desirable that such models should be equally applicable to short and continuous releases. This section describes new empirical models, collectively referred to as SGAS, that draw on features of both the FARMLAND and Nuclear Electric models. They are based on the physical processes known to be important in the uptake of sulphur by plants and draw on relevant experimental data when considering transfer rates to represent these processes. However, the magnitude of these transfer rates is also governed by two requirements. These are the need to achieve reasonable estimates of the activity concentration in the edible parts of the plant at harvest for a continuous release and reasonable predictions of the partitioning of activity in the various parts of the plant as a function of time, following a short duration release. For this reason it was necessary to develop two versions of the SGAS model, one for crops grown for their leaves and one for crops where tubers, fruits and seeds form the edible parts. These models share a similar structure but some transfer rates are adjusted accordingly. The derivation of these parameters is described in Section 5.2.

The experimental results of Kluczewski (1986) and Collins (1996A; 1996B), summarised in Section 2, are some of the more useful sources of information that have been found. These give an insight into the uptake and transfer of sulphur by plants. Other studies on the uptake of $CO^{35}S$ have shown that the rate of uptake of ^{35}S is largely dependent on resistances encountered at the air-leaf boundary and observed losses of ^{35}S from foliage arise mainly from translocation to roots and possibly from volatilisation (Coughtrey, 1983; Kluczewski, 1983). However, the data are limited and include such variability that in many cases it is only possible to make generalisations about the processes involved. The new SGAS models and transfer processes are discussed below.

5.1 The SGAS models

For modelling purposes two categories of crops were initially identified. Those for which translocation of sulphur from inedible to edible parts of the plant is important (ie grain, fruit and root vegetables) and those for which it is not (ie green vegetables and pasture). The application of the translocation model to grain and fruit gives rise to activity concentrations that are relatively high because of their smaller yields. However, the lack of data such as that available for root vegetables from Kluczewski (1983) means it is difficult to know whether these concentrations are reasonable. Nevertheless, significant transfers of activity to the edible parts of grain were observed by Collins (1996A; 1996B), see Section 2.2, and similar transfers to fruit might be expected. The application of the translocation model is described below (Section 5.1.2) only for root vegetables but its use for grain and fruit is discussed.

5.1.1 A model for leafy green vegetation

This model is closely based on a model developed by Nuclear Electric called SULPHUR (Pearce, 1991; Pearce, 1994) but it attempts to predict the activity concentrations in leafy green vegetables and pasture following both continuous and short duration atmospheric releases (Figure 1a and 1b). The edible parts of the plant are represented by compartments 1, 2, 5, 9, 10 and 11.

The rate constants used in the model are given in Tables 3 and 4 and represent those processes described in Section 5.2. The transfer from leaf to roots is set to $5\ 10^{-7}\ s^{-1}$ for the first ten days after the initial deposit to represent the fast translocation of sulphur to the plant roots. After this period it is reduced to $1\ 10^{-8}\ s^{-1}$. The higher value is used whenever deposition is occurring and therefore is used continuously in conditions where the deposition is continuous. The value of $5\ 10^{-7}\ s^{-1}$ is less than the rate derived from Chadwick (1977) because of the need to give reasonable predictions of the activity in the plant at harvest following a continuous release (Section 5.2.6). Sulphur that is deposited on the soil is modelled independently of the sulphur taken up by the leaves. This enables equilibrium to be established between the soil and the plant using the soil to plant transfer factor. If the leafy green vegetation model is used to represent pasture then the additional compartments shown in Figure 1b are used to model the undisturbed soil, instead of compartments 4 and 5 in Figure 1a. It is recommended that processing losses due to preparation of the food are not included because the remaining surface contamination is considered unimportant compared to the uptake of material across the leaf surface which is subsequently fixed by the plant.

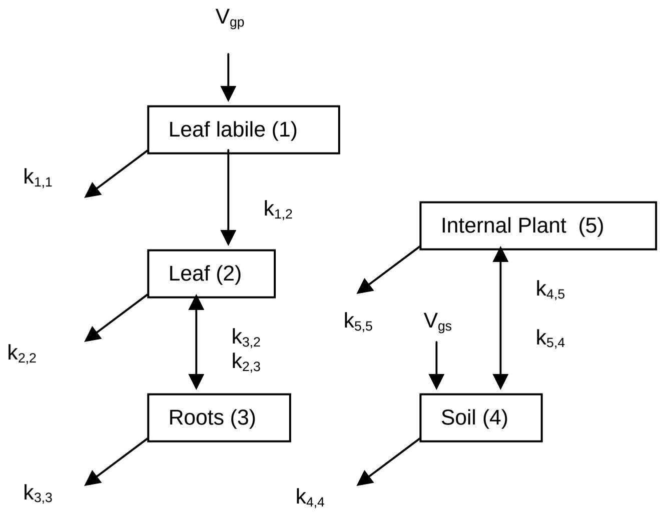

Figure 1a SGAS leafy green vegetation model

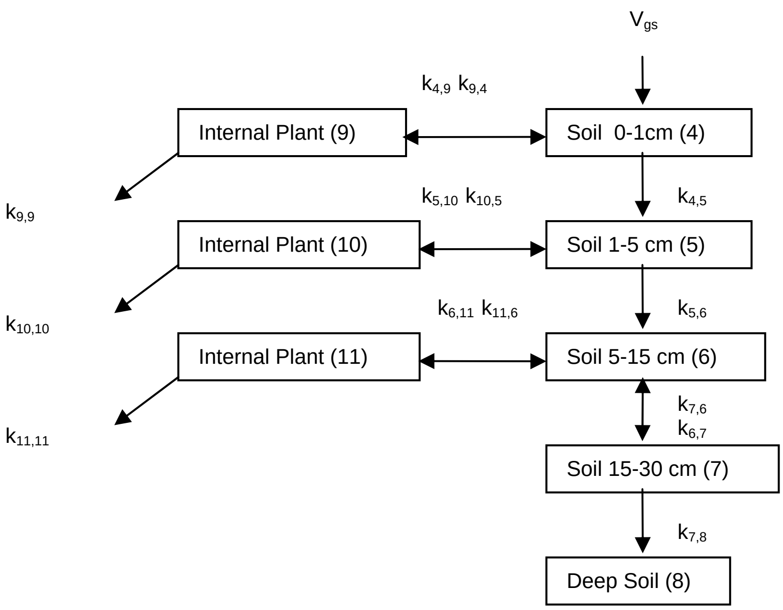

Figure 1b Additional/alternative compartments for SGAS pasture model which replace compartments 4 and 5 in Figure 1a

Table 3 Rate constants and deposition velocities for SGAS leafy green vegetation[1] model

Name	$CO^{35}S$ [2]	Units
V_{gp}	$4\ 10^{-4}$	$m\ s^{-1}$
V_{gs}	$6\ 10^{-6}$	$m\ s^{-1}$
$K_{1,1}$ (loss to atmosphere)	$2\ 10^{-2}$	s^{-1}
$K_{1,2}$ (fixed by plant)	$8\ 10^{-2}$	s^{-1}
$k_{2,2}$, $k_{3,3}$, $k_{5,5}$ (2 crops per year)	$6.3\ 10^{-8}$	s^{-1}
$K_{2,3}$ (translocation leaf to root)	$5\ 10^{-7}$ [3]	s^{-1}
$K_{3,2}$ (translocation root to leaf)	$1\ 10^{-8}$	s^{-1}
$K_{4,4}$ (soil migration- FARMLAND)	$2.2\ 10^{-10}$	s^{-1}
$K_{4,5}$ (FARMLAND)	$1.3\ 10^{-3}$	s^{-1}
$k_{5,4}$ (FARMLAND)	1	s^{-1}

1 Includes green vegetables (yield = $1\ 10^{6}$ kg km^{-2}) and pasture (yield = $5\ 10^{5}$ kg km^{-2}).

2 For $H_2{}^{35}S$ rate constants are the same as those for $CO^{35}S$ except V_{gp} is $4\ 10^{-3}$ m s^{-1}.

3 Changes to $1\ 10^{-8}$ s^{-1} ten days after deposition ceases.

Table 4 Additional/changed rate constants for SGAS pasture model

Name [1]	Rate constants (s^{-1})
	$CO^{35}S$ [2]
$k_{2,2}$, $k_{9,9}$, $k_{10,10}$, $k_{11,11}$ (grazing)	Cattle $6.0\ 10^{-7}$ Sheep $8.7\ 10^{-8}$
$K_{4,9}$ (FARMLAND)	$2\ 10^{-2}$
$K_{5,10}$ (FARMLAND)	$5\ 10^{-3}$
$K_{6,11}$ (FARMLAND)	$2\ 10^{-3}$
$K_{4,5}$ (FARMLAND)	$7.7\ 10^{-9}$
$K_{5,6}$ (FARMLAND)	$2.0\ 10^{-9}$
$K_{6,7}$ (FARMLAND)	$1.2\ 10^{-9}$
$K_{7,6}$ (FARMLAND)	$4.7\ 10^{-11}$
$K_{7,8}$ (FARMLAND)	$4.4\ 10^{-10}$

1 $k_{5,5}$, $k_{4,4}$, $k_{3,3}$ and $k_{5,4}$ are set to zero.

2 For $H_2{}^{35}S$ rate constants are the same as those for $CO^{35}S$.

5.1.2 A model for crops where translocation is important

This model is also closely based on a model developed by Nuclear Electric called SULPHUR (Pearce, 1991; Pearce, 1994) and attempts to predict the activity concentrations following both continuous and short duration atmospheric releases in root crops (Figure 2). The edible parts of the plant are represented by compartments 5, 6 and 8. Compartment 3, roots, represents those parts of the plant which are inedible and to which translocation from the leaves occurs. This includes any parts of the root system and plant stems that are not eaten.

The rate constants used in the model are given in Table 5 and represent those processes described in Section 5.2. The transfer from leaf to roots is set to 1.2 10^{-6} s^{-1} for the first ten days after the initial deposit to represent the fast translocation of sulphur to the plant roots. After this period it is reduced to 1 10^{-8} s^{-1}. The higher value is used whenever deposition is occurring and therefore is used continuously in conditions where the deposition is continuous. The value of 1.2 10^{-6} s^{-1} is similar to the rate derived from Chadwick (1977) (see Section 5.2.6). Sulphur that is deposited on the soil is modelled independently of the sulphur taken up by the leaves. This enables equilibrium to be established between the soil and the plant using the soil to plant transfer factor. The transfer factors represented by dashed lines are included for root vegetables only when harvesting occurs over a prolonged period with some immediately consumed and some stored for later consumption. This practice is referred to here as “storage” and is modelled following short duration releases when more detailed agricultural practices are needed to improve estimates of activity concentrations in root crops as a function of time. The balance box is introduced to ensure there is no reduction in activity concentration of the tuber compartments due to the transfer to storage. For grain and fruit the “storage” box is not considered as harvesting is assumed to be instantaneous. The appropriate yields for these crops (Table 5) must be used to calculate activity concentrations but otherwise the model is the same.

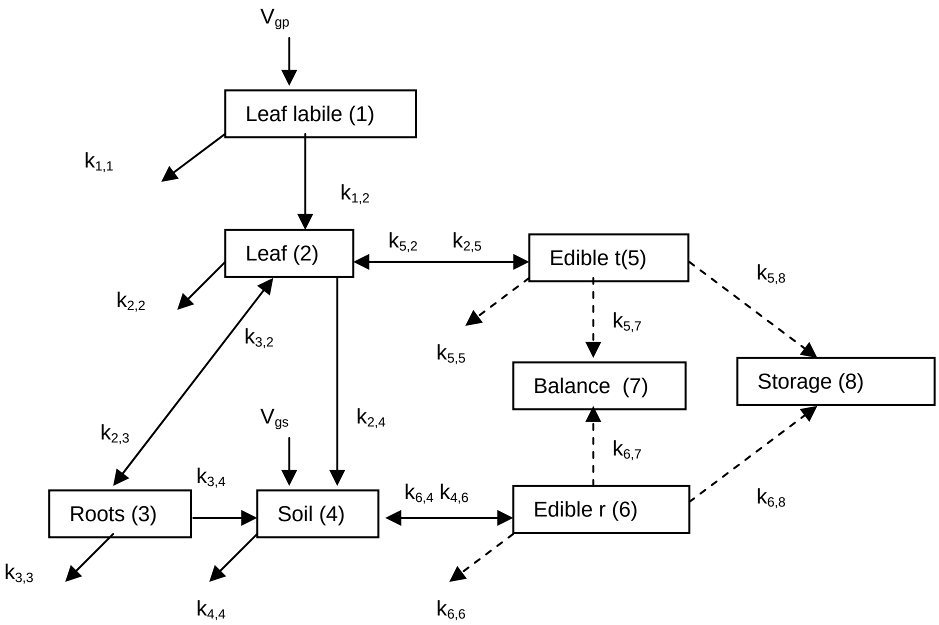

Figure 2 SGAS translocation model

Table 5 Rate constants and deposition velocities for SGAS translocation model[1]

Name	$CO^{35}S$ [2]	Units
V_{gp}	$4\ 10^{-4}$	$m\ s^{-1}$
V_{gs}	$6\ 10^{-6}$	$m\ s^{-1}$
$K_{1,1}$ (loss to atmosphere)	$2\ 10^{-2}$	s^{-1}
$K_{1,2}$ (fixed by plant)	$8\ 10^{-2}$	s^{-1}
$K_{2,2}$ (during continuous cropping only)	$3.2\ 10^{-8}$	s^{-1}
$K_{3,3}$ (during continuous cropping only)	$3.2\ 10^{-8}$	s^{-1}
$K_{2,3}$ (translocation leaf to root)	$1.2\ 10^{-6}$ [3]	s^{-1}
$K_{2,4}$ (dying back of leaves) (ploughing in of non-edible parts at start of fallow period)	$9.6\ 10^{-8}$ 1	s^{-1}
$K_{2,5}$ (translocation to edible)	$3\ 10^{-7}$	s^{-1}
$K_{3,4}$ (ploughing in of non-edible parts at start of fallow period)	1	s^{-1}
$K_{5,2}$ (translocation edible to leaf)	$1\ 10^{-8}$	s^{-1}
$k_{5,5}$, $k_{6,6}$ (cropping)	$3\ 10^{-8}$	s^{-1}
$K_{3,2}$ (translocation root to leaf)	$1\ 10^{-8}$	s^{-1}
$K_{4,4}$ (soil migration- FARMLAND)	$2\ 10^{-10}$	s^{-1}
$K_{6,4}$ (FARMLAND)	1	s^{-1}
$K_{4,6}$ (root uptake- FARMLAND)	$4.0\ 10^{-3}$	s^{-1}
$k_{5,7}$, $k_{6,7}$ (FARMLAND)	$-1.1\ 10^{-7}$	s^{-1}
$k_{5,8}$, $k_{6,8}$ (FARMLAND)	$1.1\ 10^{-7}$	s^{-1}

1 Includes root vegetables (yield = $3\ 10^{6}$ kg km^{-2}), grain (yield = $4\ 10^{5}$ kg km^{-2}) and orchard fruit (yield = $1.69\ 10^{6}$ kg km^{-2}).

2 For $H_2{}^{35}S$ rate constants are the same as those for $CO^{35}S$ except V_{gp} is $4\ 10^{-3}$ m s^{-1}.

3 Changes to $1\ 10^{-8}$ s^{-1} ten days after deposition ceases.

5.2 Transfer processes

5.2.1 Foliar absorption

Foliar uptake through the stomata in the leaf surface is the principal pathway for the uptake of $CO^{35}S$ and $H_2{}^{35}S$ gases by plants. Therefore, environmental conditions that affect the action of the stomata will also influence the uptake rate of these gases. For example, in conditions of high light intensity and plentiful water supply the stomata will be fully open and gaseous uptake will be high. Conversely, in drought conditions and at night the uptake of gases will be greatly reduced. This uptake mechanism can be represented in a model by using a deposition velocity.

5.2.2 Deposition velocities to plants and uptake by leaves

In the models described below it is assumed that only dry deposition occurs which is reasonable given the low solubility of the compounds considered ($CO^{35}S$ ~3.5 g l^{-1} and $H_2{}^{35}S$ ~7.0 g l^{-1} at 0°C and 1 bar). This is in contrast for example to SO_2 which is highly soluble (~228 g l^{-1} at 0°C and 1 bar). Therefore, when modelling the dispersion of $CO^{35}S$

and $H_2{}^{35}S$ under meteorological conditions which include rain a washout coefficient of zero would be used. There are a significant amount of data in the literature that give experimentally determined values of the dry deposition velocity of $CO^{35}S$ and $H_2{}^{35}S$ on to plants. For gaseous forms of ^{35}S the deposition velocity is a measure of the total amount of airborne activity intercepted by the plant and includes uptake by leaves. Deposition velocities have been reviewed by NRPB and can be found in Smith (1998). The default values recommended for use with this model are $4\ 10^{-4}$ m s^{-1} for $CO^{35}S$ and $4\ 10^{-3}$ m s^{-1} for $H_2{}^{35}S$ to plants.

5.2.3 Deposition velocities to soils

The low solubility of $CO^{35}S$ and $H_2{}^{35}S$ may explain the small deposition velocities obtained for soils (Kluczewski; 1983). The default value recommended for use here is $6\ 10^{-6}$ m s^{-1} for $CO^{35}S$ and $H_2{}^{35}S$. These values are considerably less than those for deposition onto plants where the active uptake of gaseous ^{35}S across the leaf is important.

5.2.4 Leaf labile

Data on the loss of ^{35}S from plants is limited. Recent studies (Collins, 1996A; Collins, 1996B) suggest that the only loss mechanism from plants exposed to aerosols of $CO^{35}S$ and $H_2{}^{35}S$ is radioactive decay. However, other researchers have measured sulphur-containing gases emanating from plants. In Coughtry (1983) losses from plant leaves of the order of 15% to 25% are quoted. It has been suggested (Stewart, 1999) that these losses might occur from labile pools of sulphur. Within these pools sulphur becomes fixed over time and hence losses to the atmosphere gradually reduce. In this study the models include a labile leaf compartment within which sulphur is fixed by the plant ($k_{1,2} = 8\ 10^{-2}\ s^{-1}$) and lost to atmosphere ($k_{1,1} = 2\ 10^{-2}\ s^{-1}$) in a ratio of 4 to 1.

5.2.5 Root uptake

Uptake of $CO^{35}S$ and $H_2{}^{35}S$ through the plant root system is much less important, following an atmospheric release, than foliar uptake due to the small deposition velocity of these gases onto soils. Therefore, the transfer from soil to plant has been estimated using the method previously adopted in FARMLAND. A soil to plant equilibrium concentration ratio is used and it is assumed that rapid equilibrium is reached between the plant and the soil. Root uptake only contributes significantly to the activity concentration in the plant when the major source of contamination is to the soil compartment. In a review of the behaviour of sulphur in terrestrial ecosystems (Coughtrey, 1983) a dry weight plant to dry weight soil concentration ratio for radioactive isotopes of sulphur of about 10 was recommended for most conditions. When converted to wet mass plant to dry mass soil the concentration ratio is a factor of 1. A review of soil to plant concentration factors has been carried out in Brown (1995) and recommends a value of 0.6 which is used here. Soil to plant transfer factors calculated using the following approach are presented in Table 6.

Concentration ratio	=	Bq kg^{-1} wet weight plant / Bq kg^{-1} dry weight soil
Fraction of activity transferred from soil to plant in unit time	=	(wet mass plant / dry mass soil) x Fraction of activity transferred from plant to soil in unit time x concentration ratio

Table 6 Root uptake rate constants used in SGAS models for different plant types

Plant type	Yield (kg km^{-2})	Root uptake (s^{-1})
Leafy green vegetables (For soil depth 0 to 30 cm)	$1\ 10^{6}$	$1.3\ 10^{-3}$
Pasture grass (For soil depth 0 to 1 cm)	$5\ 10^{5}$	$2.0\ 10^{-2}$
Pasture grass (For soil depth 1 to 5 cm)	$5\ 10^{5}$	$5.0\ 10^{-3}$
Pasture grass (For soil depth 5 to 15 cm)	$5\ 10^{5}$	$2.0\ 10^{-3}$
Root vegetables (For soil depth 0 to 30 cm)	$3\ 10^{6}$	$4.0\ 10^{-3}$

1 Dry mass of soil per unit area = $1.5\ 10^{7}$ kg km^{-2} per cm depth

2 Fraction of activity transferred from plant to soil in unit time = 1 s^{-1} (to reach equilibrium quickly)

5.2.6 Translocation between foliage and roots

There is evidence to suggest (Chadwick, 1977) that the field loss of $CO^{35}S$ from grass is initially rapid with a half-life of about 6.7 days for the first 10 days following deposition (ie a loss of about 65% in 10 days) but is then much slower. If it is pessimistically assumed that the only loss mechanism is translocation to the roots this gives a leaf to root transfer factor of $1.2\ 10^{-6}$ s^{-1}. The same study showed the rate of loss of $H_2{}^{35}S$ to be much slower, having a half-life of about 25 days (ie a loss of about 25% in 10 days). However, other studies (Collins, 1996A; Collins, 1996B)) found the same initial rapid transfer from leaves to roots for both $CO^{35}S$ and $H_2{}^{35}S$. Coughtrey (1983) gives evidence to suggest that as much as 25% of foliar-absorbed sulphur can be expected to be translocated down to the roots in fumigated plants. It has also been shown that in perennial rye grass up to 30% of ^{35}S can be translocated to the roots within 24 hours of fumigation with $CO^{35}S$ (Kluczewski, 1983). In the leafy green vegetation model this process has been modelled using a transfer factor of $5\ 10^{-7}$ s^{-1}. This corresponds to about 40% transfer from leaves to roots over 10 days. It is somewhat slower than the value derived from (Chadwick, 1977) for $CO^{35}S$ but it enables reasonable predictions to be made in both short and continuous release applications. For the translocation model a value of $1.2\ 10^{-6}$ s^{-1} is used which is based on the data from Chadwick as described above. The long term component ($1\ 10^{-8}$ s^{-1}) has been chosen to achieve reasonable predictions of activity concentrations in the edible parts of plants following a short duration release.

A pessimistic estimate of the rate at which material is returned from the roots to the plant has been made. Coughtrey (1983) reported that the fraction of ^{35}S applied to soil that is subsequently translocated upwards from the roots of barley is between 1.7% to 11.8%. If there is no foliar uptake ($k_{2,3} = 0$) then at equilibrium it can be shown that:

$$\frac{N_2}{N_3} = \frac{k_{3,2}}{k_r}$$

where

N_x = Number of atoms m^{-2} in compartment x.

N_2/N_3 = concentration factor between foliage and root, $CF_{r.f}$

k_r = radioactive decay rate

Therefore

$$k_{3,2} = k_r CF_{r.f}$$

If 0.12 is taken to be a pessimistic estimate of $CF_{r.f}$ then $k_{3,2}$ is estimated to be about 1.1 10^{-8} s^{-1}.

5.2.7 Translocation to edible tissue

This term represents the transfer of sulphur from the parts of the plant not normally eaten to the seeds or tubers that are consumed. It is therefore an important process that needs to be considered when modelling uptake of material by crops such as potatoes. It is essentially the same process as that described in Section 5.2.6 except that material is being transferred to a part of the plant that is growing rapidly. As a result it is reasonable to assume that transfer to these edible parts will be greater in the longer term than transfer to those parts of the plant that are growing less. This transfer factor has been selected on the basis that it gives a reasonable estimate of the activity concentrations as a function of time following a short release but also at harvest (assuming a 120 day growing period) following a continuous release. The data for short releases are based on experiments carried out with grain, beans and potatoes (Section 2.2). However, for continuous releases activity concentrations at harvest are only available for root vegetables and beans (Section 2.1). The value of the transfer factor used to model this process is 3 10^{-7} s^{-1} and represents a half-time of about 25 days. Consequently, the amount of deposited activity that is transferred to edible tissue in the first 10 days is about 14%. This increases to about 35% after 4 months. There is evidence to suggest that this transfer may initially be higher because significant fractions of sulphur taken up through the leaves of plants have been found in tubers and seeds.

5.2.8 Loss from foliage to soil

The main process modelled here is the loss which arises as a consequence of the dying back of foliage. It has been suggested that this can be ignored for leafy vegetables such as cabbage but might be included for other crops such as potatoes. It is approximated as 1/(length of the growing season). With a growing season of 120 days this term is estimated to be 9.6 10^{-8} s^{-1}.

5.2.9 Transfer from root to soil

The lack of data available on this process means that it has not been included in the model but it is likely to be a relatively unimportant transfer mechanism.

5.2.10 Resuspension of soil to plant surface

This contamination route has not been included because it is thought to be insignificant in comparison to root uptake for sulphur.

6 RESULTS AND DISCUSSION

The SGAS models are based closely on the SULPHUR model developed by Nuclear Electric (Pearce, 1991; Pearce, 1994). However, where SULPHUR is only appropriate for modelling short duration atmospheric releases the SGAS models are appropriate for both short and continuous atmospheric releases. Data for both these types of release are available only for $CO^{35}S$ and hence the models have been set up based on this gas. When applied to releases of $H_2{}^{35}S$ an enhanced deposition velocity to plants, which is a factor of 10 greater than that for $CO^{35}S$, should be used. Model results, excluding losses for cropping, have been compared with results from SULPHUR and also those from the FARMLAND model to give an indication of the implications of the proposed modifications (Figures 3 to 12 and Tables 7 and 8). It should be noted, however, that problems have been encountered when trying to set up a model that is applicable to both types of release because the uptake following a short duration release appears to be much higher than that following a continuous release. This problem can only be resolved satisfactorily through further experimental studies.

Predicted activity concentrations in leafy green vegetables using SGAS are shown in Figure 3. Only a fraction of the activity initially captured by the leaves will be retained, the rest is released back to the atmosphere. There is an initial rapid transfer of the retained activity from the leaves to the roots over the first ten days following exposure. This is something that has been noticed by many researchers. There then follows a period of reduced translocation where the activity remains within the leaves (edible tissue) and roots and any loss of activity from these compartments is due entirely to radioactive decay. The partitioning of the activity is shown in Figure 4. After the initial rapid transfer to roots approximately 60% of the sulphur that was fixed by the plant remains in the leaves while the rest is in the roots. This is broadly consistent with the findings of Collins (1996A; 1996B) that are summarised in Table 2. As time passes the activity in both leaves and roots tends to 50% of the total in the plant. This model also fits the data for pasture grass reasonably well (Pearce, 1991) and, if combined with the standard FARMLAND models for grazing animals, could be used to predict activity concentrations in animal products such as lamb, beef and milk. For comparison the same results are presented for the SULPHUR ‘leaf’ model in Figures 5 and 6. In this model deposition of activity occurs onto plant leaves and soil and, once activity has been fixed by the leaf, the only losses are due to radioactive decay. Consequently, the total activity in the SULPHUR model is higher than that in SGAS. SULPHUR does not change the value of transfers after 10 days as is done in SGAS but instead has transfers from leaf to root and root to leaf that have half lives of about 7 days and 3 days respectively. These give the desired long term leaf to root ratio shown in Figure 6. A comparison of SGAS with FARMLAND is shown in Figure 12 for a short duration release. For leafy green vegetables FARMLAND predictions are comparable with those of SGAS for the first 10 days but subsequently decrease more rapidly due to more significant losses, namely losses from plant to soil and from the soil out of the system.

Predicted activity concentrations in root vegetables using SGAS are shown in Figure 7. Again, only a fraction of the activity initially captured by the leaves will be retained, the rest is released back to the atmosphere. There is an initial rapid transfer of activity from the leaves to the roots over the first ten days following exposure. In addition, there is a steady transfer of sulphur from the leaves to the edible parts of the plants (tubers) which continues at a fixed rate. The partitioning of the activity is shown in Figure 8. After the initial rapid transfer to roots there is a steady build up of activity in the edible parts of the plant i.e. the tubers. It is possible that this occurs at an even faster rate but it has been difficult to model this while at the same time ensuring the model adequately predicts the activity concentration in the tuber at harvest (assuming a 120 day growing period) following a continuous release. As time passes the activity in the tubers tends to about 60% of the total activity fixed by the plant. Experimental data from Collins (1996A; 1996B) suggests that about 50% of the total activity in the plant is recovered from the tubers (Table 2). Similar fractions of the total initial activity were also observed in the edible parts of other crops such as grain. The same results are also presented for the SULPHUR 'pulse' model in Figures 9 and 10. The main differences between the models are that SULPHUR predicts more activity in the edible tissue and leaf and less in the roots and soil. The amount of activity transferred to the edible parts in SGAS is constrained by the data for continuous releases and consequently more activity is moved to the roots and soil. In contrast SULPHUR does not have this constraint and models transfer from leaf to roots and leaf to edible tissue at the same rate. It is possible that SGAS does not move enough activity to the edible parts of the plant in the first few days following deposition but the experimental data vary significantly and suggest that this transfer process depends on the stage of growth of the plant. However, SULPHUR predicts activity concentrations in root vegetables that are only a factor of two greater than those from SGAS. A comparison of SGAS with FARMLAND is shown in Figure 12 for a short duration release. This shows that FARMLAND predictions are comparable with those of SGAS for the first 30 days but subsequently decrease more rapidly due to more significant losses, namely losses from the edible part of the plant to soil and from the soil out of the system

It should be noted that the activity concentrations shown in Figures 11 and 12 (and Tables 7 and 8) are for unit air concentration for a continuous release, and unit integrated air concentration for a short duration release. In each case the model has been run assuming there are no losses due to cropping or grazing. It can be seen that for a continuous release the activity concentrations predicted by FARMLAND for leafy green vegetables, pasture and root crops are similar to those predicted by the SGAS models for $CO^{35}S$. However, for short duration releases the SGAS $CO^{35}S$ models predict significantly greater levels of activity in the leaves of crops and this has greatest impact on activity concentrations in green vegetables and pasture. This difference is about a factor of 10 at 60 days after exposure. Because of the difference in the value of the deposition velocity used between FARMLAND (1 10^{-3} m s^{-1}) and the SGAS $CO^{35}S$ (4 10^{-4} m s^{-1}) models the difference in the activity concentration per unit deposition will be even more significant. If the results are based on activity concentrations per unit deposit the predictions from the SGAS $CO^{35}S$ model will be systematically (1 10^{-3}/ 4 10^{-4}) 2.5 times higher than the differences shown here and can be compared with the data in Section 2.2.

The application of the SGAS $CO^{35}S$ model to short duration releases can have a significant impact on the dose assessment. The model predictions in Figure 8 and Table 8 can be divided by the deposition velocity for $CO^{35}S$ to give activity concentrations per unit deposition. The peak values can be compared with those bounding values in Section 2.2 that were derived from measurements. They are broadly similar with the greatest difference being for root vegetables where model predictions are a factor of about 4 less than the values derived from measurement.

For $H_2{}^{35}S$ a deposition velocity to plants of 4 10^{-3} m s^{-1} is recommended for use in the SGAS models. However, for all other parts of the models the same rate constants that are recommended for $CO^{35}S$ have been used. This seems justified given the variability in the data. Consequently, the activity concentrations per unit deposit are very similar between $CO^{35}S$ and $H_2{}^{35}S$, but $H_2{}^{35}S$ activity concentrations per unit air concentration are higher by a factor of ten.

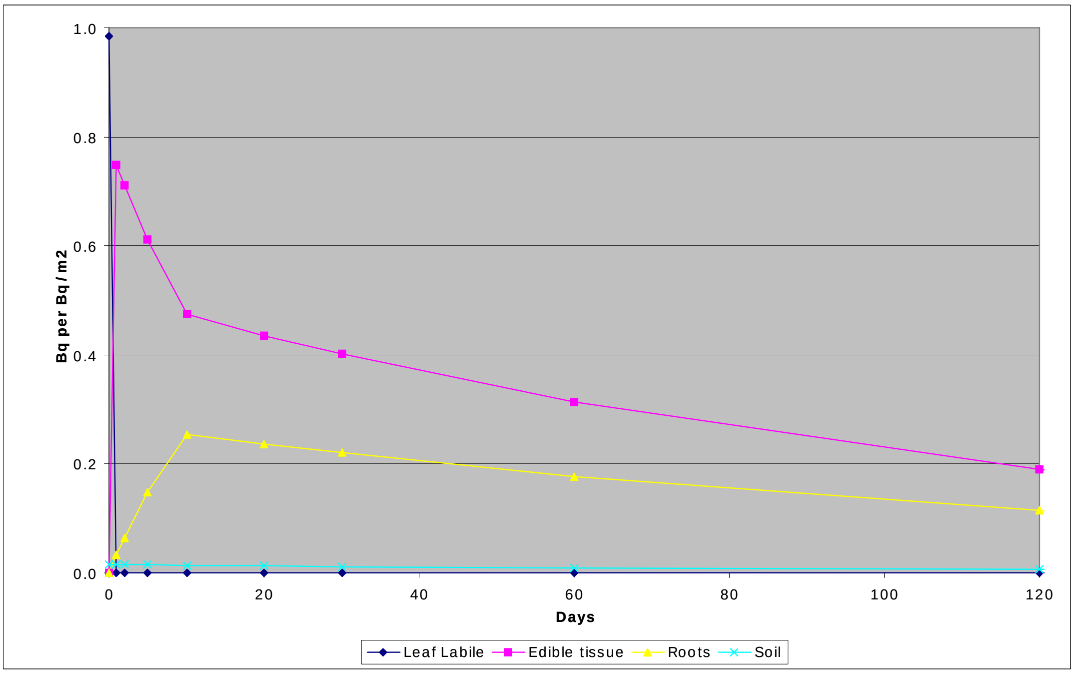

Figure 3 Activity in leafy green vegetables following unit deposition from spike release (Bq/Bq m^{-2}) using SGAS $CO^{35}S$ model

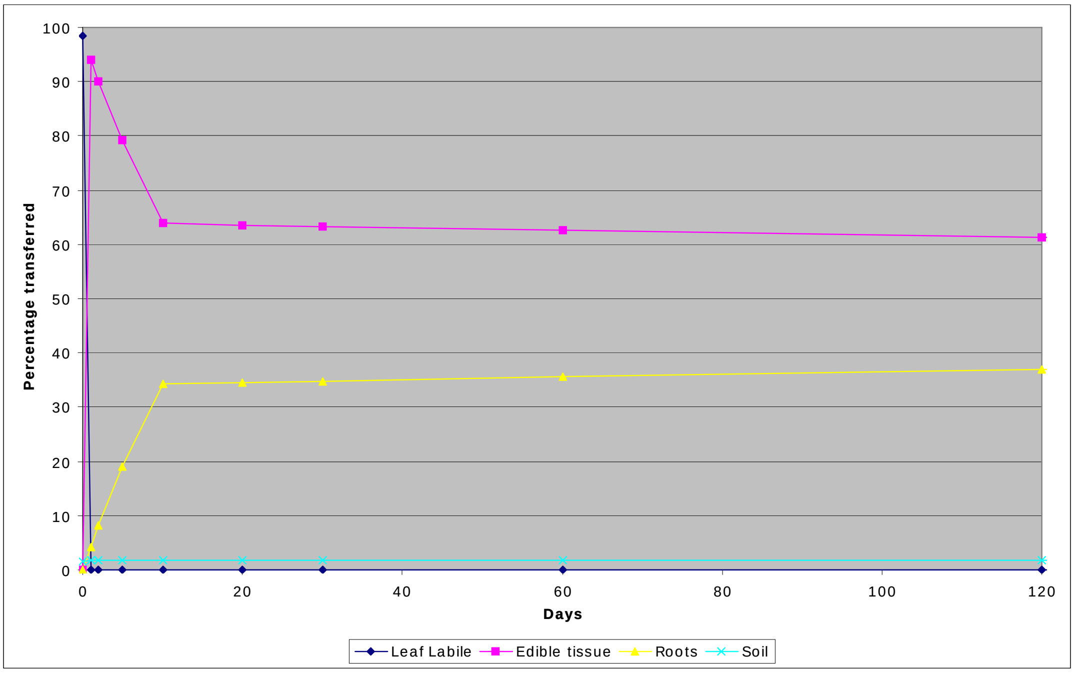

Figure 4 Partitioning of activity in leafy vegetables following unit deposition from a spike release using SGAS $CO^{35}S$ model

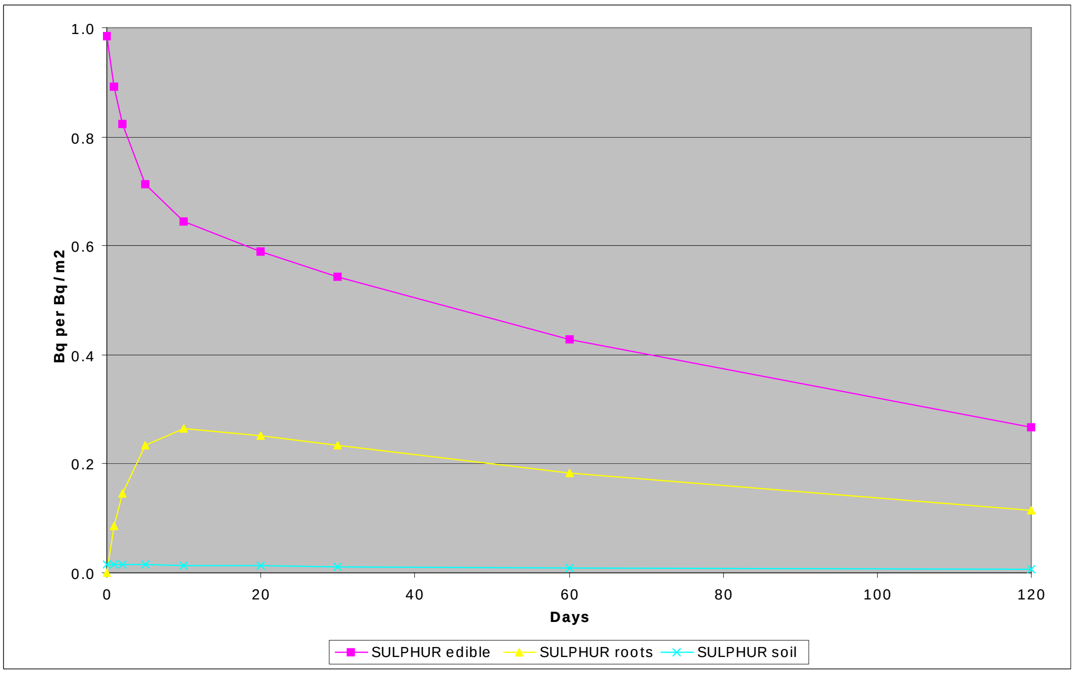

Figure 5 Activity in leafy green vegetables following unit deposition from spike release (Bq/Bq m^{-2}) using SULPHUR $CO^{35}S$ model

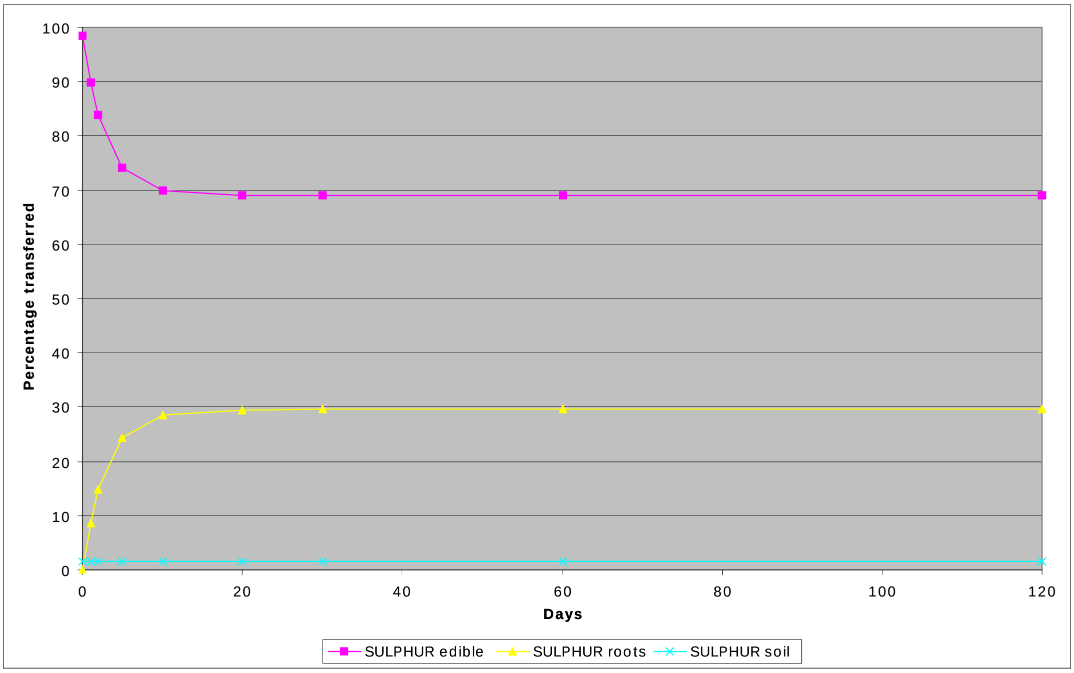

Figure 6 Partitioning of activity in leafy vegetables following unit deposition from a spike release using SULPHUR $CO^{35}S$ model

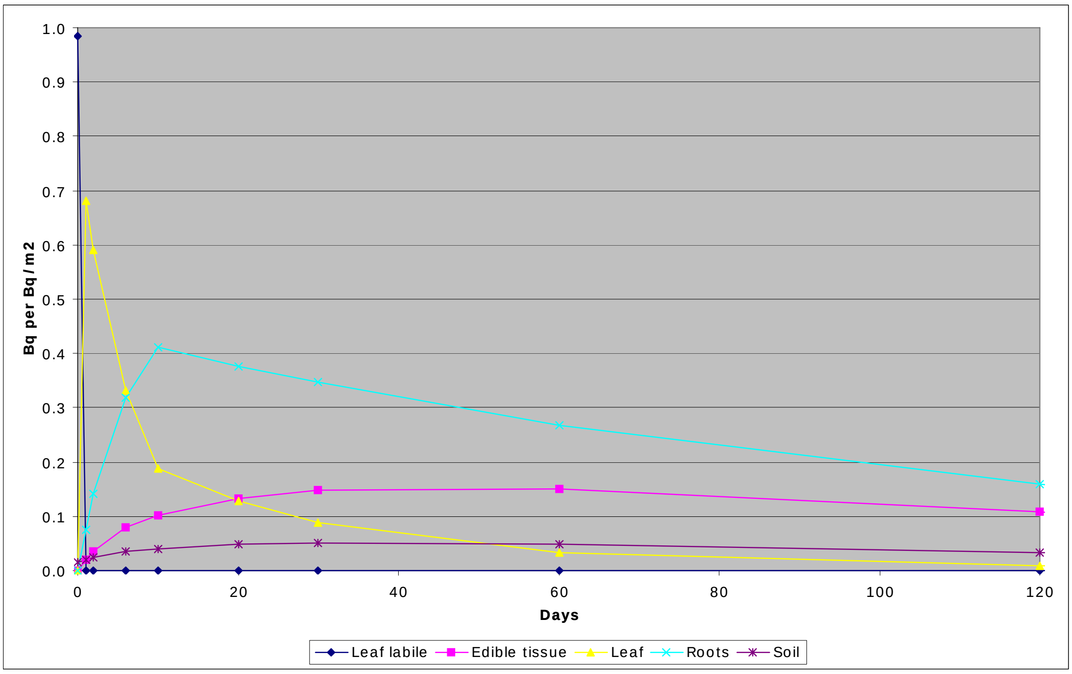

Figure 7 Activity in root vegetables following unit deposition from a spike release (Bq/Bq m^{-2}) using SGAS $CO^{35}S$ model

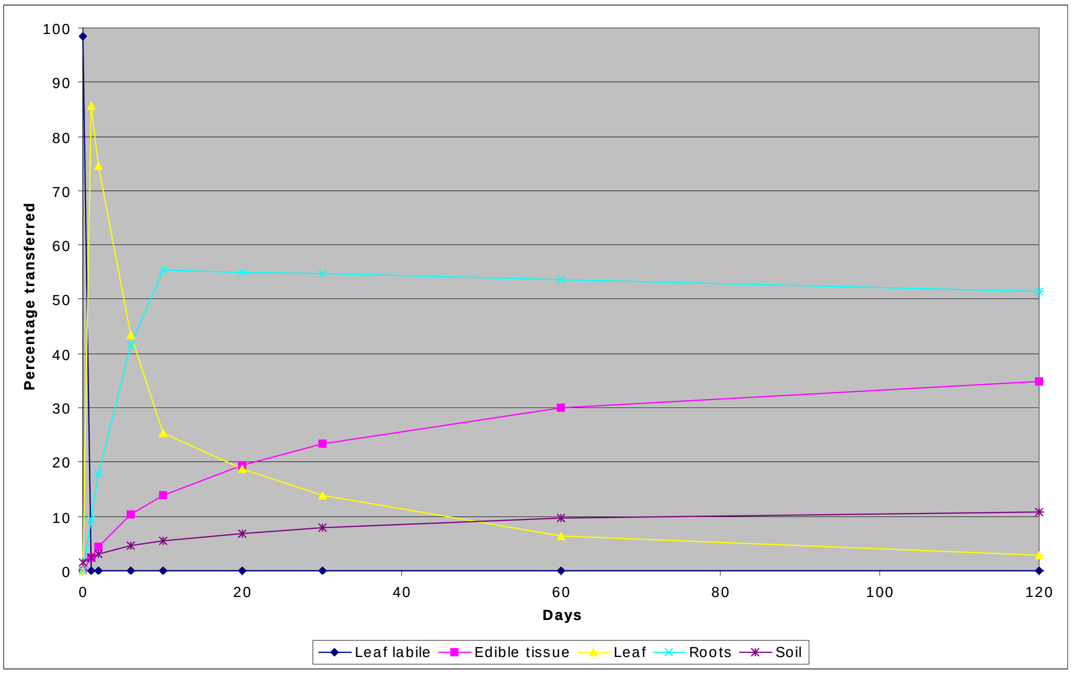

Figure 8 Partitioning of activity in root vegetables following unit deposition from a spike release using SGAS $CO^{35}S$ model

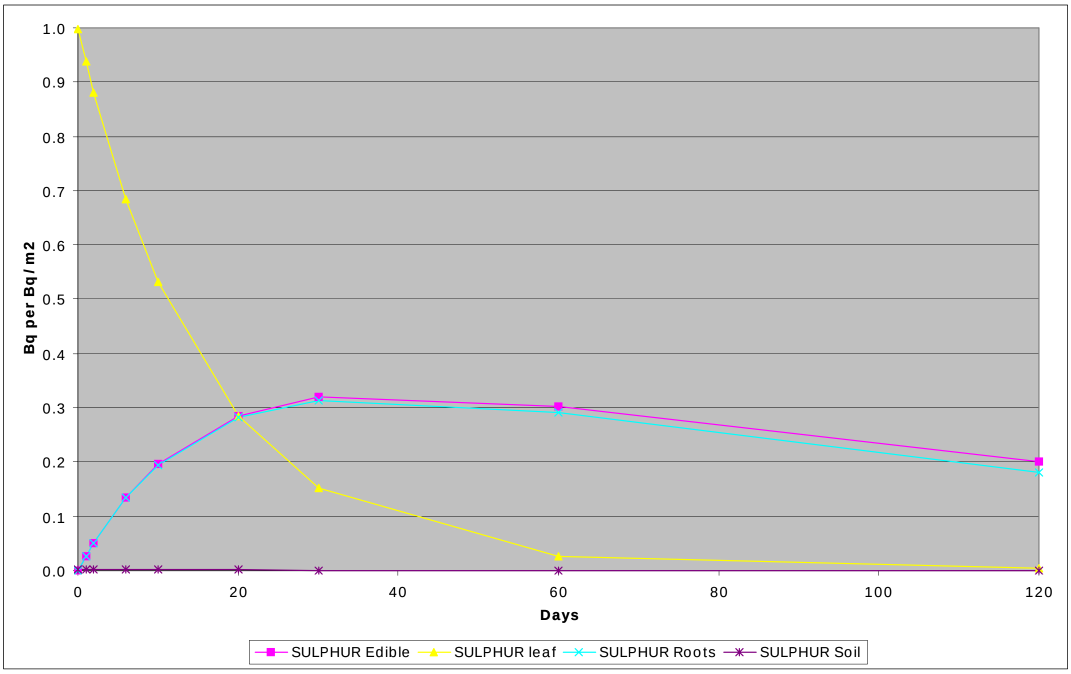

Figure 9 Activity in root vegetables following unit deposition from a spike release (Bq/Bq m^{-2}) using SULPHUR $CO^{35}S$ model

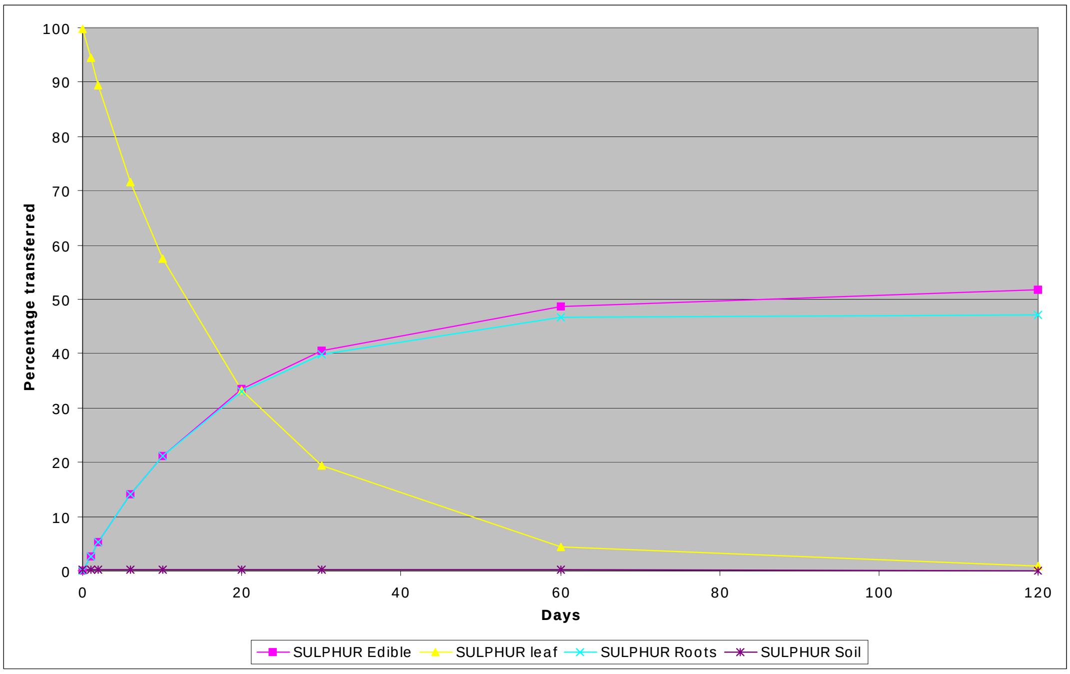

Figure 10 Partitioning of activity in root vegetables following unit deposition from a spike release using SULPHUR $CO^{35}S$ model

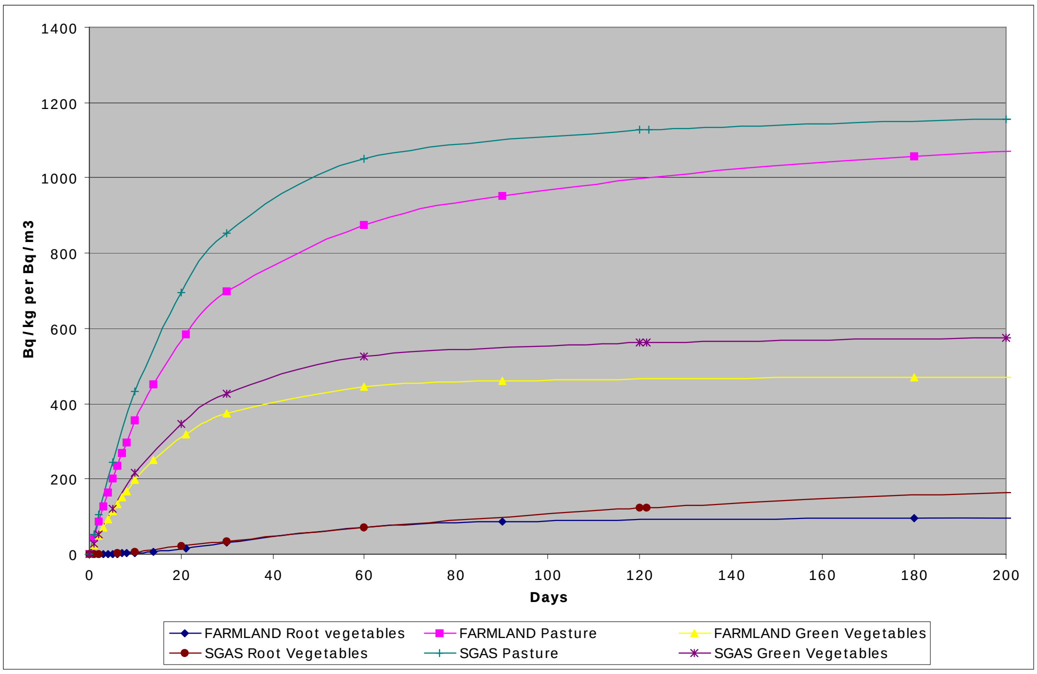

Figure 11 Activity concentrations in crops per unit air concentration (Bq kg^{-1}/Bq m^{-3}). FARMLAND and SGAS $CO^{35}S$ models with continuous release

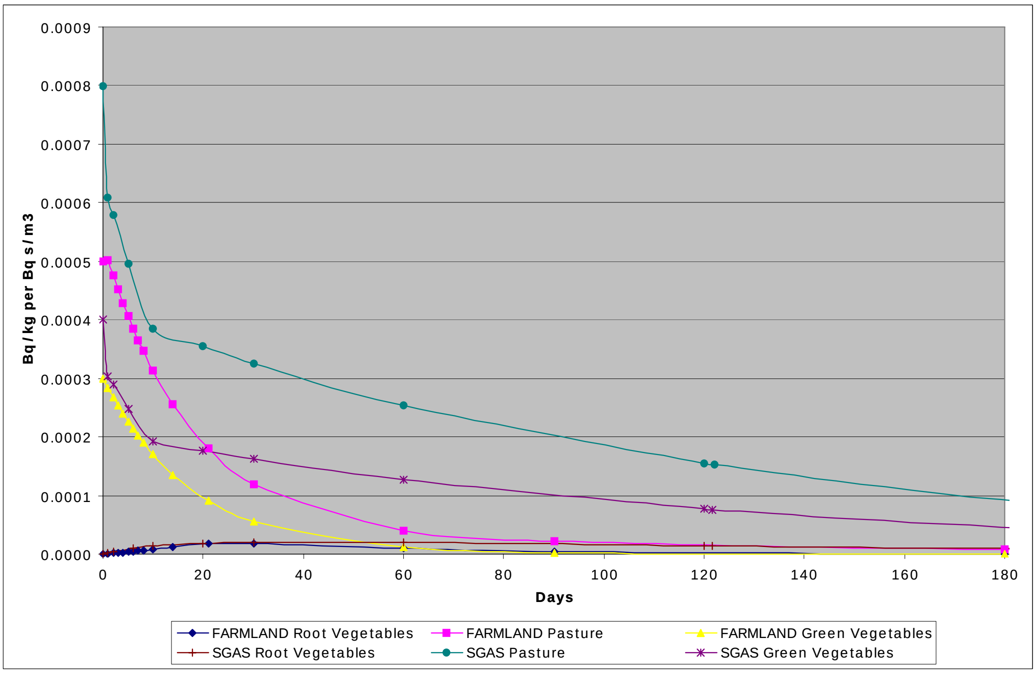

Figure 12 Activity concentrations in crops per unit integrated air concentration (Bq kg^{-1}/Bq s m^{-3}). FARMLAND and SGAS CO^{35}S models with spike release

Table 7 Activity concentrations in crops at harvest (120 days) for a continuous release of CO^{35}S to atmosphere

Plant	Activity Concentration (Bq kg^{-1} per Bq m^{-3})	
	FARMLAND	SGAS CO^{35}S models
Green vegetables	460	563
Root vegetables	90	123
Pasture	1000	1130

Table 8 Activity concentrations in crops in the first 30 days following a spike release of $CO^{35}S$ to atmosphere

Plant	Times since exposure (days)	Activity Concentration (Bq kg^{-1} per Bq s m^{-3})	
		FARMLAND	SGAS $CO^{35}S$ models
Green vegetables	0	3.00 10^{-4}	4.00 10^{-4}
	1	2.84 10^{-4}	3.04 10^{-4}
	2	2.68 10^{-4}	2.89 10^{-4}
	5	2.26 10^{-4}	2.48 10^{-4}
	10	1.70 10^{-4}	1.92 10^{-4}
	30	5.55 10^{-5}	1.63 10^{-4}
Root vegetables	0	0	0
	1	9.68 10^{-7}	2.57 10^{-6}
	2	1.40 10^{-6}	4.77 10^{-6}
	5	3.71 10^{-6}	1.08 10^{-5}
	10	8.88 10^{-6}	1.39 10^{-5}
	30	1.88 10^{-5}	2.00 10^{-5}
Pasture	0	5.00 10^{-4}	8.00 10^{-4}
	1	5.02 10^{-4}	6.08 10^{-4}
	2	4.76 10^{-4}	5.78 10^{-4}
	5	4.06 10^{-4}	4.96 10^{-4}
	10	3.13 10^{-4}	3.85 10^{-4}
	30	1.18 10^{-4}	3.26 10^{-4}

7 IRRIGATION AND SEWAGE

The SGAS models shown in Figures 1 and 2 are based on data derived from experimental studies of the uptake and transfer of $CO^{35}S$ and $H_2{}^{35}S$ following atmospheric releases. If sulphur is applied to agricultural land by spray irrigation or the application of sewage sludge then the uptake mechanisms will be very different. Due to the lack of data it was decided to adopt the approach previously used in FARMLAND to model root uptake. The performance of the SGAS models following irrigation and the application of sewage sludge has been investigated by comparison with the existing FARMLAND model. Activity concentrations in leafy green vegetables, root vegetables and soil following continuous irrigation at 100 Bq m^{-2} y^{-1} have been estimated and are shown in Figure 13. It should be noted that losses due to cropping, which are the same for both sets of models, have not been included and therefore the activity concentrations in soil under green vegetables will in fact be lower than those shown in Figure 13. In addition, activity concentrations in pasture and soil following continuous sewage application at a rate of 1 Bq m^{-2} y^{-1} are shown in Figure 14.

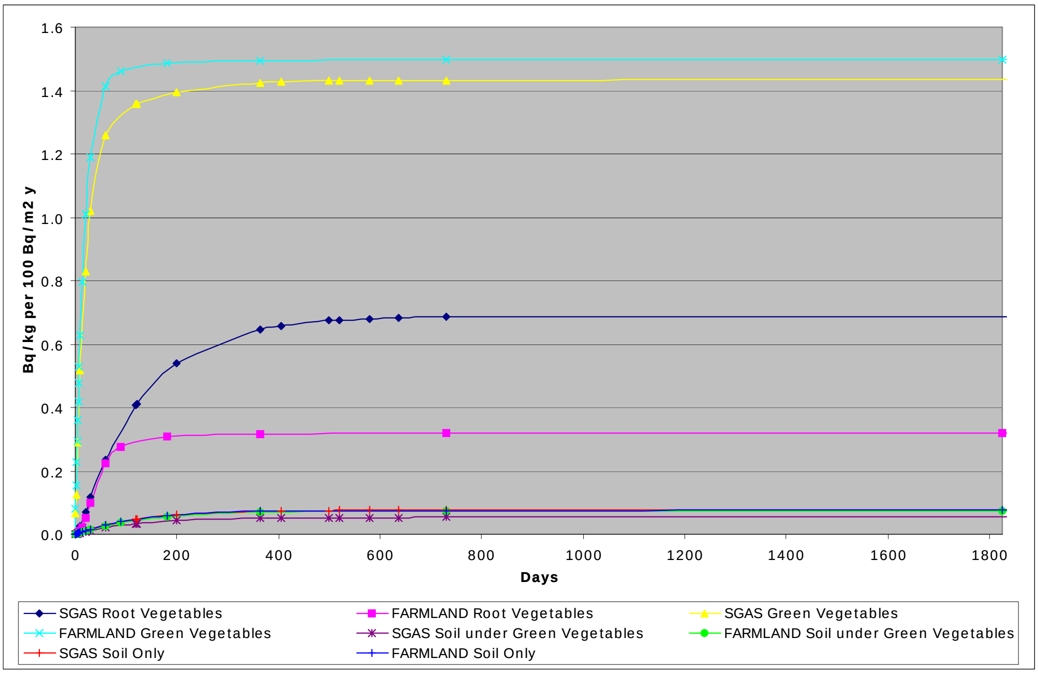

Figure 13 Activity concentrations in crops following irrigation (Bq kg^{-1} per 100 Bq m^{-2} y^{-1}). FARMLAND and SGAS CO^{35}S models with continuous release

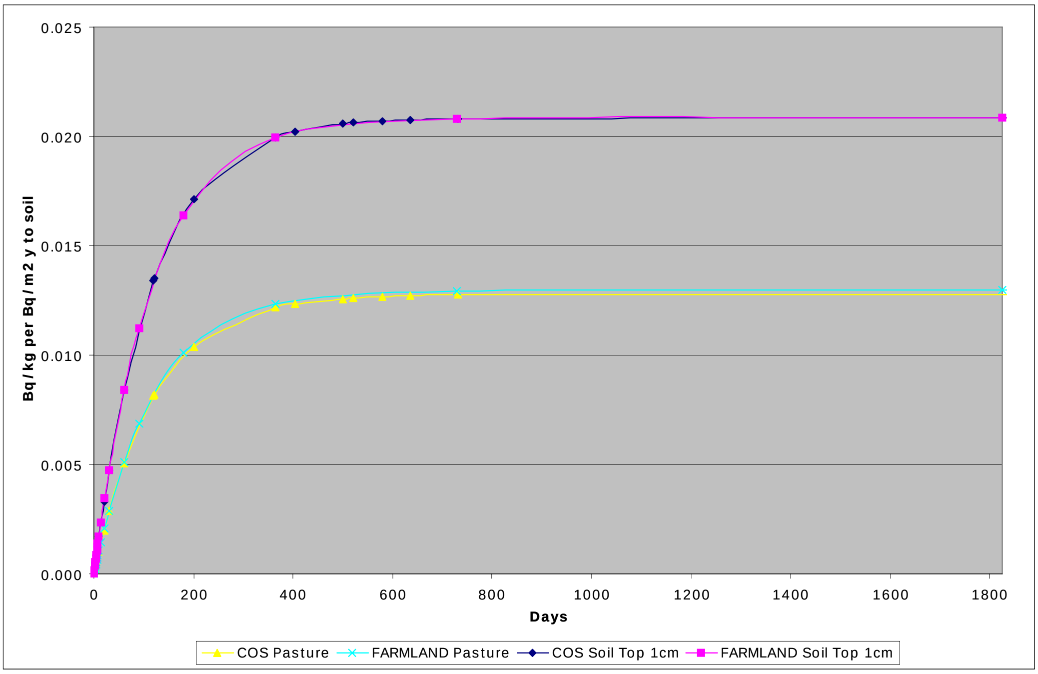

Figure 14 Activity concentrations in crops following sewage application to soil (Bq kg^{-1} per Bq m^{-2} y^{-1}). FARMLAND and SGAS CO^{35}S models with continuous release

Following spray irrigation it is assumed that a certain fraction (30%) of the water is deposited directly onto the plant while the rest deposits on to the soil. This mechanism of contamination replaces the deposition velocity approach described for gaseous releases. Therefore, uptake via both the plant leaves and roots is important. The most noticeable differences arise in the activity concentrations of root vegetables and these are attributable to the different ways in which translocation has been modelled (Figure 13). The SGAS $CO^{35}S$ model includes a higher translocation rate in an attempt to predict the high levels of activity that have been found in the edible parts of root vegetables immediately after a short duration atmospheric release.

It is assumed that sewage sludge is applied directly to the soil and that there is no interception by the plant. The soil to plant concentration ratios used in the two models are the same and hence the activity concentrations in pasture at equilibrium are very similar (Figure 14).

Generalised Derived Limits (GDLs) are levels of radioactivity in environmental media that are likely to give rise to a dose of 1 mSv to members of the public (NRPB, 2000). They are calculated for single radionuclides and for exposure pathways relevant to the contaminated media. The calculation of GDLs requires the activity concentration in the appropriate environmental media in the 50th year from 50 years of discharge. The SGAS $CO^{35}S$ sulphur models have been used as input to the calculation of GDLs for sulphur in sewage sludge and irrigation water by estimating the exposure from ingestion of food produced from conditioned soil. In all cases the models described here predict these quantities within a factor of about two of the FARMLAND models.

8 RECOMMENDATIONS AND FUTURE WORK

The FARMLAND model is not considered suitable for application to short duration planned releases of gaseous sulphur. It is therefore recommended that the SGAS models described in this report or the SULPHUR models developed by Nuclear Electric are used instead. The SGAS models may also be used for continuous release scenarios. However, these models should only be used as screening tools given the many uncertainties associated with them. It is also suggested that, in the absence of better information, the SGAS translocation model may be used as a screening tool to assess the uptake of $CO^{35}S$ and $H_2{}^{35}S$ by grain and fruit following short or continuous releases.

There are a number of areas that would benefit from further experimental study such as the determination of appropriate deposition velocities. A wide range of deposition velocities has been measured and values have been shown to depend on environmental factors such as light intensity and availability of water. It would be useful to investigate further the dependence of deposition velocity on these factors and also on the plant species.

Although a very useful source of data one of the problems with Collins (1996A; 1996B) is that the transfer of sulphur through the plant was estimated by analysing different plants at different times after fumigation. This is why, in some cases, there is an apparent increase in total activity in the plant with time. The variability in the data makes it difficult to determine trends that might provide insight to the processes affecting the transfer of sulphur in plants. This will always be a problem because of the destructive measurement techniques involved. However, if sample sizes could be increased then it may be possible to identify trends.

The Collins study was also carried out against a background air concentration that was sulphur free. It would be interesting to study plant response to elevated levels of $CO^{35}S$ and $H_2{}^{35}S$ where background air concentrations of sulphur are closer to those measured in the field.

It would be useful to study the uptake and partitioning of sulphur by various crops following continuous fumigation in the laboratory. This would enable apparent differences in the behaviour of sulphur following short fumigations in the laboratory and continuous fumigation in the field to be resolved. In addition, further investigation of the mechanisms by which sulphur is lost from plants particularly the foliage would be of interest. The role of labile pools of sulphur should be considered.

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