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PardNor –  
PARAmeters for ingestion Dose models  
for NORdic areas -  
Status report for the NKS-B activity 2010

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## Abstract

The ECOSYS foodchain model is built into the European standard decision support systems ARGOS and RODOS, which are integrated in the preparedness for radiological events in the Nordic countries. However, a review has revealed that a number of parameters in ECOSYS do not reflect the current state-of-the-art knowledge, and do not adequately represent Nordic conditions. Improved and country/region specific data is required for ECOSYS to give trustworthy results. It is the aim of the PardNor activity to collect new data, and thus enable reliable use of ECOSYS for scenarios involving contamination of Nordic food production areas. In the reported work period of the PardNor activity, the parameters governing the contaminant deposition processes were revised, and an important point here is that contaminant particle sizes were taken into account, which has so far not been the case in ECOSYS. Both dry and wet deposition processes were addressed. New datasets were derived for dry deposition, whereas for wet deposition (washout, rainout, snow scavenging), which can not be addressed directly in ECOSYS, but must be dealt with elsewhere in the ARGOS and RODOS decision support systems, a new methodology was suggested on the basis of available measurement data. Also parameters governing the natural weathering processes of contaminants on crops and bare soil were revised, and it was demonstrated that precipitation has a strong influence on the weathering half-life, which should be included in ECOSYS. Both for deposition and weathering parameters, a special effort was made to retrieve measurement data of Nordic origin. A series of calculations were made with the ECOSYS model to show the effect of introducing new and improved parameter values for dry deposition and weathering processes. The parameter revision was found to have great effect on the ECOSYS estimates of food contamination levels for a 'Chernobyl-like' NPP accident scenario, and the effect could well be even greater for other conceivable types of release scenarios. Finally, the dependence of measured global fallout contamination levels on precipitation rates was highlighted in a separate section.

## Key words

Foodchain modelling, ingestion dose, ECOSYS, transfer factors, radioactive contamination

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# **PardNor**

## **PARameters for ingestion Dose models for NORdic areas**

Status report for the NKS-B activity 2010

**Edited by Sven P. Nielsen and Kasper G. Andersson**

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# 1. Introduction

In compliance with the agreement with NKS, this year's work on the PardNor activity had two main objectives, both directed towards improving the Nordic knowledge platform required to make reliable estimates, using the ECOSYS model, of ingestion doses in the event of a contaminating incident. As the ECOSYS model is implemented in both of the European decision support systems, ARGOS and RODOS, which are used by the Nordic authorities, the work constitutes an important step towards an improved and harmonised Nordic preparedness. The two main tasks this year were:

(i) To improve the modelling of the contaminant deposition process to agricultural crops as well as to bare soil. The primary improvement lies in the implementation of particle size dependent deposition parameters. The ECOSYS model has so far operated with particle size independent parameterisation, which is problematic and can lead to considerable calculation errors. Both parameters describing the dry and wet deposition processes were revised, although wet scavenging parameters can not be implemented in ECOSYS, but must be entered elsewhere in the European decision support systems that ECOSYS is a part of. An additional effort was made to derive deposition velocity data from measurements made specifically in Nordic areas. A series of ECOSYS model runs with plausible scenarios were made to investigate the effect of the suggested parametric changes.

(ii) To improve the parameterisation of natural weathering processes for contaminants deposited on agricultural crops. The more specific objectives identified by the work group were here: (a) to improve weathering rates, taking into account the host of data that has been made available since the ECOSYS model parameterisation was last revised, (b) to investigate the influence of rainfall on the weathering process, with a view to improving ECOSYS prognoses by including weather data here, (c) to derive weathering data from measurements made specifically in Nordic areas, and (d) to run ECOSYS for a series of plausible scenarios to determine the effect of suggested weathering parameterisation changes.

With specific reference to the modelling of wet deposition processes, a discussion was added on a study of the relationship between recorded precipitation rates and global fallout deposition densities.

In 2010, a journal paper was published on work done within the PardNor activity: Hansen, H.S., Nielsen, S.P., Andersson, K.G., Thørring, H., Joensen, H.P., Isaksson, M., Kostianen, E., Suolanen, V., Sigurgeirsson, M.A. & Pálsson, S.E. (2010). Effect of Nordic diets on ECOSYS model predictions of ingestion doses, *Radiation Protection Dosimetry* 140 (2): pp. 182-190.

Also, a paper on PardNor work was presented at the Third European IRPA Congress, Helsinki, Finland, 14-18 June, 2010, and will appear in the congress proceedings: Andersson, K.G., Nielsen, S.P., Thørring, H., Hansen, H.S., Joensen, H.P., Isaksson, M., Kostianen, E., Suolanen, V. & Pálsson, S.E. (2010). Improving ingestion dose modelling for the ARGOS and RODOS decision support systems: A Nordic initiative.

Finally, a PardNor activity related abstract has been accepted for oral presentation at the International Conference on Radioecology & Environmental Radioactivity, Hamilton, Ontario, Canada, 19-24 June 2011: Andersson, K.G., Nielsen, S.P., Thørring, H., Hansen, H.S., Joensen, H.P., Isaksson, M., Kostianen, E., Suolonen, V. & Pálsson, S.E. Parametric improvement for the ingestion dose module of the European ARGOS and RODOS decision support systems.



## 2. Improvement of deposition velocity estimates

The default values in ECOSYS for dry deposition velocities to crops have no published reference to any study, and thus seem to emerge as an ‘educated guess’ (Müller & Pröhl, 1993), demonstrating that little information of direct relevance was available at the time when the defaults were formed. It is claimed by Müller & Pröhl that the data relates to an aerosol size distribution in the range of 0.1-1  $\mu\text{m}$ , but the same values are used as defaults for all aerosol contaminants. Nonetheless, since different elements and their pre-release compounds have different physicochemical properties, they will undergo different aerosolisation processes in connection with a release involving an explosion or fire. Depending on the release process, some of the contaminants will evaporate and form small condensation particles, whereas other will be released in the form of somewhat larger fragmentation particles. For instance in connection with the Chernobyl accident, it was observed that the different contaminant aerosol that contaminated large food producing areas could be distinguished in two groups: a volatile group, including  $^{132}\text{Te}$ ,  $^{134}\text{Cs}$ ,  $^{137}\text{Cs}$ ,  $^{99}\text{Mo}$ ,  $^{103}\text{Ru}$ , and  $^{106}\text{Ru}$ , with an AMAD (Activity Median Aerodynamic Diameter) of the order of slightly less than 1  $\mu\text{m}$ , and a ‘refractory’ group, to which  $^{140}\text{Ba}$ ,  $^{95}\text{Zr}$ ,  $^{141}\text{Ce}$ ,  $^{144}\text{Ce}$ ,  $^{89}\text{Sr}$ , and  $^{90}\text{Sr}$  belong, with higher AMAD values of the order of 4  $\mu\text{m}$  (Andersson et al., 2002). This means that the ECOSYS defaults should be particularly appropriate for the first of these two groups. However, as the effect of gravitational settling would be quite pronounced at a particle size of 4  $\mu\text{m}$  (Slinn, 1982), these particles would in general have a considerably higher deposition velocity, as was also observed on various surfaces following the Chernobyl accident (e.g., Roed, 1990). It is therefore necessary to distinguish between deposition velocities for different particle size groups, as has also in recent years been done in connection with the modelling of the urban contaminant deposition process in the ARGOS and RODOS decision support systems (Andersson et al., 2008).

Although a number of studies have been conducted over the years of dry deposition to vegetation, the data that is directly applicable to improve ECOSYS parameterisation for agricultural crops is rather sparse. The majority of studies seem to relate to non-radioactive gaseous atmospheric releases (e.g.,  $\text{SO}_2$ ,  $\text{O}_3$ ,  $\text{HNO}_3$ ,  $\text{NH}_3$ ), whereas some studies of relevant contaminants have too little detail in the air pollutant characterisation to be useful, and either completely omit information on aerosol size (e.g., McLeod et al., 1984; Pinder et al., 1990), or refer only to an aerosol size range, such as  $\text{PM}_{10}$  (e.g., Mitchell et al., 2010). Further, particle sizes are in other studies too large to be relevant at all for widespread airborne dispersion. Other studies only give deposition velocities to a single crop leaf rather than to the plant as a whole (e.g., Singhal et al., 2004; Wedding et al., 1975), and in yet other cases, air sampling was clearly not operated isokinetically, wherefore the results cannot be used (e.g., Ferrandino & Aylor, 1985). In early iodine experiments (Sehmel, 1980), the chemical form of the iodine is often undefined or known to be of a mixed nature (e.g., of  $\text{HIO}_3$ ,  $\text{HIO}_4$  and organic iodides), and results thus of little or no value.

Table 2.1 gives an overview of some experimentally obtained values for the deposition velocity parameter. From what has been specified in the literature, these values seem to reflect typically prevalent atmospheric and environmental conditions. The parameters might under given ‘untypical’ circumstances have values that would not be well represented by these figures. Some figures have here been left out of the table, where wind velocities were very high and this clearly affected the deposition

velocity. It has been demonstrated (Ahmed, 1979) that between wind velocities of 2 and 14 m s<sup>-1</sup>, the deposition velocity of naturally occurring radioactive aerosols increases by about a factor of 3, both to smooth (e.g., filter paper) and rough (grass) surfaces. In-line with this, it has also been shown (Freer-Smith et al., 2003; Slinn, 1982) that deposition velocities of 0.8 µm particles to vegetation (trees) can increase by a factor of 3-4 between wind velocities of 3-9 m s<sup>-1</sup>. As the particle size increases beyond about 20 µm, the influence of wind speed on deposition increases markedly, due to the significance of the inertial impaction mechanism (Ahmadi & Li, 1999). However, such large particles will in any case only remain airborne for short time, due to their large mass, and radionuclides associated with these would thus only contaminate rather small areas, depending on, e.g., the initial plume rise height. Currently, the interface structures of the ARGOS and RODOS systems do not allow consideration of the influence of wind speed on deposition, although wind speed data already used in the system could in the future be linked to deposition parameters through semi-empirical formulae. Deposition velocity also depends on atmospheric stability. It has been demonstrated that under moderately stable atmospheric conditions (e.g., night time with clear sky), the friction velocity,  $u_*$ , will only be about half of its value under neutral conditions (Jensen, 1981). This in turn means that the eddy diffusion part of the deposition velocity will be reduced to about a quarter (IAEA, 1994) (this is excluding the sedimentation contribution to the deposition velocity).

Also surface roughness is an important parameter influencing the deposition velocity. An indication of this influence can be seen from measurements made in the Roskilde area after the Chernobyl accident. Here <sup>137</sup>Cs deposition velocities to grassed surfaces varied rather widely (Roed, 1990) between 1.8 and 8.8 m s<sup>-1</sup>. However, if the length of the grass is taken into account (by dividing with the grass mass per unit area), the results are consistent within 10 %. The division by grass mass per unit area also gave consistent data for elemental iodine deposition velocities, which varied similarly. The effect of crop/grass growth with time is included in ECOSYS through the LAI parameter (many different crops actually have very similar maximum LAI; Nielsen & Andersson, 2008). It is however also very important to consider the length of mature grass at the time and place in question (could be very long in a permanent pasture, and subject to regular cutting in other types of areas).

Table 2.1. Measured deposition velocities of aerosols and iodine gases to a range of agricultural crops.

Crop type	V <sub>d</sub> (Iodine gas), mm/s	V <sub>d</sub> (aerosol), mm/s	Aerosol AMAD, µm	Crop LAI****	Reference
Head lettuce	0.29*	0.11	0.7	Mature plants	Tschiersch et al. (2009)
Leafy lettuce (summer)	0.37*	0.14	0.7	8.1 (5)	Tschiersch et al. (2009)
Lettuce	-	1.7	4	Mature plants	Watterson & Nicholson (1996)
Lettuce	-	4.6	10	Mature plants	Watterson & Nicholson (1996)

Lettuce	-	27	22	Mature plants	Watterson & Nicholson (1996)
Wheat	-	1.3	4	Mature plants	Nicholson & Watterson (1992)
Wheat	-	46	22	Mature plants	Nicholson & Watterson (1992)
Wheat (dry)	-	36	30	Mature plants	Jonas (1984)
Cereals	-	19-78	24	Mature plants	Chamberlain & Chadwick (1972)
Cereals	-	30	30	Mature plants	Chamberlain (1975)
Spinach (summer)	1.6*	0.08	1.1	4.7 (-)	Tschiersch et al. (2009)
Curly kale	0.89*	0.26	1.1	Mature plants	Tschiersch et al. (2009)
White cabbage	0.16*	0.02	1.1	Mature plants	Tschiersch et al. (2009)
Endive	0.38*	0.11	0.7	8.2 (-)	Tschiersch et al. (2009)
Short grass (ca. 0.2 kg/m <sup>2</sup> )	2.2*	0.4	0.7	Cut	Roed (1990)
Short grass	-	0.8	4	Cut	Roed (1990)
Grass	5.5*	-	-	Unclipped	Cline et al. (1965)
Grass	4-11*	-	-	-	Vogt (1974)
Grass	-	30	19	-	Chamberlain (1967)
Longer grass (ca. 1 kg/m <sup>2</sup> )	9*	0.7	0.7	Nature area	Roed (1990)
Longer grass (ca. 10-15 cm)		0.7	0.5	Nature area	Bonka (1989)
Longer grass (ca. 10-15 cm)		0.5	0.7	Nature area	Clark & Smith (1988)
Moss		80	24	-	Clough (1975)
Cabbage	0.003**	-	-	Mature plants	Collins et al. (2004)
Bean	0.002**	-	-	Mature plants	Collins et al. (2004)
Carrot	0.001**	-	-	Mature plants	Collins et al. (2004)
Pasture grass	0.001-0.02**	-	-	-	Atkins (1967)

\* Elemental iodine gas

\*\* Methyl iodine gas

\*\*\* ECOSYS default in brackets for comparison

Table 2.1 demonstrates the strong dependence of deposition velocity on aerosol size. For instance between aerosol sizes of ca. 4  $\mu\text{m}$  and ca. 20  $\mu\text{m}$ , the deposition velocity is seen to increase by about a factor of some 20-30. The data in most cases show only rather limited influence of the crop species (also between crops as different as cereal plants and lettuce investigated by the same workers), although it is clear that there is some variation that merits further experimental investigations to further improve model reliability. However, although leaf areas and structures of the fully developed plants can of course vary somewhat between species (Nielsen & Andersson, 2009; Müller & Pröhl, 1993), the above directly relevant data does not provide safe grounds for introducing different values for different crops. If the ARGOS/RODOS developers so wish, it may however be possible in the future to improve on this by taking into account the substantial experimental data for deposition of other gases as well as that for deposition to non-edible plants with structural similarities. It is important to note that the figures originating from the study by Tschiersch et al. (2009) are in most cases low, compared with values measured for other plants, both for aerosols and elemental iodine gas (Chamberlain, 1967; Garland, 2001; Jonas & Vogt, 1982; Jonas, 1984; Petroff, 2005; Roed, 1987; Roed, 1990). The study by Tschiersch et al was a small scale chamber experiment with wind conditions (e.g., very low wind velocity) that do not represent field conditions well, and as also noted by the author, the *absolute* values should therefore be used with caution, whereas *relations* of deposition velocity between species and contaminant categories should be in better agreement with those that would be observed under field conditions.

For all crops, except grass and fruit, the ECOSYS default is for aerosols 2 mm/s, for elemental iodine gas 20 mm/s, and for organic iodine gas 0.2 mm/s. Table 2.2 compares these defaults with a set of crop type blind values that could be derived from the data in Table 2.1 (applying the highest reported values from the experiment by Tschiersch et al., to reasonably comply with the Chernobyl data for other vegetation mentioned above as well as with theoretical considerations). Values are given for the particle size groups and iodine gases typically reported for the Chernobyl accident, as well as for a group of large airborne particles that could still be dispersed over kilometres, e.g., by an extensive and prolonged fire (Danish Atomic Energy Commission (1970). This shows that the ECOSYS aerosol defaults are actually most suitable for the 4  $\mu\text{m}$  group, contrary to what was stated, and ECOSYS defaults are too high by a factor of ca. 4-10 for the other contaminant groups.

Table 2.2. Comparison between ECOSYS default deposition velocities to agricultural crops and a new dataset derived from the data in Table 2.1.

Dataset	$V_d$ (1 $\mu\text{m}$ aerosol), mm/s	$V_d$ (4 $\mu\text{m}$ aerosol), mm/s	$V_d$ (20 $\mu\text{m}$ aerosol), mm/s	$V_d$ (elemental iodine gas), mm/s	$V_d$ (organic iodine gas), mm/s
This study	0.3	1.5	35	5	0.02
ECOSYS defaults	2	2	-	20	0.2

As for grass, the default for aerosols was 1.5 mm/s, and for elemental and organic iodine it was 15 mm/s and 0.15 mm/s respectively. Also these values seem somewhat too high, even for pasture grass, when compared with the values in Table 2.1. Little is known of the deposition velocity to fruit. The ECOSYS defaults suggest that this is

in general higher than the deposition velocities to other crops by a factor of 2.5. In lack of better information, it may be suggested to apply the values in Table 2.2 multiplied by a factor of 2.5 for fruit.

Of course, during seasons with no standing crops (bare soil) or snow covered land, deposition velocities in crop production areas are quite different. The ECOSYS default deposition velocity to bare soil is 0.5 mm/s for aerosols, 3 mm/s for elemental iodine, and 0.05 mm/s for organic bound iodine. However, on the basis of the literature, it is suggested to use a value of 0.1 mm/s for aerosols of the 1  $\mu\text{m}$  range, 0.4 mm/s for aerosols of the 4  $\mu\text{m}$  range, 3 mm/s for elemental iodine, and 0.02 mm/s for organic bound iodine (Sehmel, 1980; McMahon & Denison, 1979; Bunch, 1968; Jonas & Vogt, 1982). For deposition to snow-covered soil, the corresponding deposition velocities would according to the literature be of the order of 0.6 mm/s for the 1  $\mu\text{m}$  aerosol group, 1 mm/s for the 4 $\mu\text{m}$  aerosol group, 3 mm/s for elemental iodine, and 0.02 mm/s for organic bound iodine (Sehmel, 1980; Bunch, 1968; Jonas & Vogt, 1982; Davidson et al., 1996; Davidson et al., 1981; Dovland & Eliassen, 1976; Forland & Gjessing, 1975; Ibrahim et al., 1983; Tschiersch et al., 1991).

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### 3. Improvement of washout and rainout coefficient estimates

In the past, the European decision support systems did not distinguish between contaminants with different particle sizes. This has now to some extent changed with respect to dry deposition parameterisation for *inhabited* areas, so that some of the long-distance effects of large accidents at nuclear power plants can now be represented in a reasonable way (but parameterisation for the larger particles that could arise in for instance malicious dispersion scenarios has still not been included). Also with respect to wet deposition, it is important to take into account particle size and ensure that a reasonable level of detail is applied. This section is aimed at giving an overview of how particularly particle size, but also rain rate, affects the scavenging process.

#### 3.1. Washout

Washout is the commonly used term for the process of *below-cloud* scavenging of pollutants from a contaminated plume. Due to washout, the airborne particle concentration will during rainfall at constant intensity decrease exponentially with time. The relevant time constant is called the scavenging coefficient,  $\Lambda$ , and is usually given in units of  $s^{-1}$ . The scavenging coefficient depends considerably on the size of the particles that are scavenged by the rain. However, for particle sizes greater than about 5  $\mu m$ , the scavenging process becomes independent of the particle size, since inertial impaction of aerosol particles on the rain drops very effectively governs the washout (Chate et al., 2003). For smaller particles, the influence of inertial impaction decreases, and the scavenging coefficient reaches a minimum between 0.2 and 1  $\mu m$ . For even smaller particles, the scavenging coefficient is an inverse function of the particle size, since the dominating mechanism driving the washout is here the Brownian diffusion of the particles to the scavenging rain drop (Martin et al., 1980; Wang & Pruppacher, 1980). The washout process also strongly depends on the rain rate. Tschiersch et al. (1995) demonstrated that the minimum in scavenging rate at large submicron particles is only pronounced at low to moderate rain rates (i.e., at 0.6 mm/h, to a lesser degree at 10 mm/h and almost not at all at 40 mm/h). Näslund & Holmström (1993) suggested the following semi-empirical polynomial function set to describe the dependence of the scavenging coefficient,  $\Lambda$ , on the particle radius,  $r$  ( $\mu m$ ) and the rain rate  $q$  (mm/h):

$$\begin{aligned} r < 0.5 \mu m: \Lambda &= 0 \\ 0.5 \mu m < r < 10 \mu m: \Lambda &= (b_0 + b_1 r + b_2 r^2 + b_3 r^3) f(q) \\ 10 \mu m < r: \Lambda &= f(q), \\ f(q) &= a_1 q + a_2 q^2, \end{aligned}$$

$$\text{where } a_1 = 2.7 \cdot 10^{-4}; a_2 = -3.618 \cdot 10^{-6}; b_0 = -0.1483; b_1 = 0.3220133; b_2 = -3.0062 \cdot 10^{-2}; b_3 = 9.34458 \cdot 10^{-4}.$$

However, this formula does not agree well with the experimental findings of Tschiersch et al. (1995), who demonstrated that the influence of rain rate is for the large particles ( $> 5 \mu m$ ) much less. Also Näslund & Holmström's equation set is incorrect for the smallest ( $< 0.5 \mu m$ ) particles, where Brownian diffusion results in a scavenging coefficient that is far from 0. Baklanov & Sørensen (2001) proposed to

apply the equation  $\Lambda = 8.4 \cdot 10^{-5} q^{0.79}$  for the smaller ( $r < 1.4 \mu\text{m}$ ) particle fraction, but also this is problematic, since it does not account for the strong particle size dependence.

For particle radii greater than  $1.4 \mu\text{m}$ , Baklanov & Sørensen demonstrated that the above formula set is in good agreement with the theoretical formula used by Crandall et al. (1973) and Slinn (1977):

$$\Lambda = \frac{q}{2a_m} \left[ \frac{4}{P_e} (1 + 0.4 \text{Re}^{1/2} \text{Sc}^{1/3}) + \frac{4r_p}{a_m} \left( \frac{r_p}{a_m} + \frac{(1 + 2\mu_w r_p / \mu_a a_m)}{(1 + \text{Re}^{-1/2} \mu_w / \mu_a)} \right) + \left( \frac{\rho_w}{\rho_a} \right)^{1/2} \left( \frac{\text{St} - \text{St}_*}{\text{St} - \text{St}_* + 2/3} \right)^{3/2} \right]$$

where  $q$  is the rain rate (mm/h),  $a_m$  is the volume-mean raindrop radius,  $P_e$  is the Peclet number,  $\text{Re}$  is the Reynolds number,  $\text{Sc}$  is the Schmidt number,  $r_p$  is the particle radius,  $\mu_a$  and  $\mu_w$  are the dynamic viscosities of respectively air and water,  $\rho_a$  and  $\rho_w$  are the densities of respectively air and water,  $\text{St}$  is the Stokes number, and  $\text{St}_*$  is the critical Stokes number. Since the radiocaesium aerosol from the Chernobyl accident was only about  $0.3 \mu\text{m}$  in radius (see, e.g., Andersson et al., 2002), and even smaller particles could be dispersed over a city area following a terrorist attack using a radiological dispersion device (Prouza & Hulka, 2009; Andersson et al., 2009), it is highly important to describe the scavenging coefficient adequately also for this part of the aerosol spectrum. The experimental data of Tschiersch et al. show that between about 0 and  $0.5 \mu\text{m}$ ,  $\Lambda$  can be modelled as a linearly decreasing function of the particle radius that is about  $8 \cdot 10^{-4} \text{s}^{-1}$  at a radius approaching 0, and one or two orders of magnitude less (depending on the rain rate) at the radius of about  $0.5 \mu\text{m}$ .

Also other parameters affect the aerosol capture efficiency, and thereby the scavenging coefficient. For instance, according to Tschiersch et al. (1995), the washout coefficient depends on the size distribution of the drops. If there are many large drops, the scavenging coefficient will vary less with particle size. Also electrophoresis, thermophoresis and diffusiophoresis may influence the washout scavenging coefficient, but practically only for particle sizes between  $10^{-2}$  and  $1 \mu\text{m}$  (Sparmacher et al, 1993).

Further, if the contaminant aerosols are hygroscopic, their size can increase in humid air, which will change their capture efficiency, and thereby the washout coefficient (Stensland et al., 1975). Chate et al. (2003) showed that for some hydroscopic particles (consisting of NaCl or  $(\text{NH}_4)_2\text{SO}_4$ ) smaller than  $5 \mu\text{m}$ , humidity can change the scavenging coefficient by several orders of magnitude.

### 3.2. Rainout

Rainout is the commonly used term for the process of *in-cloud* scavenging of pollutants from a contaminated plume. The convective aerosol scavenging process that occurs within a rain cloud has generally been reported to be more efficient in leading to deposition than washout (Baklanov & Sørensen, 2001), and the convective rainout coefficient does not appear to be very sensitive to changes in aerosol size (Crandall et al., 1973). Maryon & Ryall (1996) suggested the following aerosol size invariant formula for determining the convective rainout coefficient,  $\Lambda_c$  [ $\text{s}^{-1}$ ], as a

function of the rain rate,  $q$ , in mm/h:  $\Lambda_c = 3.36 \cdot 10^{-4} q^{0.79}$ . In addition, a process similar to that described above for below-cloud scavenging will occur, and this component of the scavenging coefficient can be adequately estimated using the formula described for washout (Baklanov & Sørensen, 2001).

### 3.3. Snow scavenging

The snow scavenging process is unfortunately generally not well examined and understood. Some workers (e.g., Maryon & Ryall, 1996; Hongisto, 1998; Sportisse, 2007) suggest that the use of the same model for scavenging by snow and by rain is appropriate. However, although the conceptual models and relative influences of for instance aerosol size and precipitation intensity seem to apply for both types of precipitation, the coefficient for scavenging by snow has been reported by Baklanov & Sørensen (2001) to generally be *smaller* than the washout coefficient for the same precipitation rate and aerosol size, by a factor of 2-10. Contrary to this, according to the experimental results of Sparmacher et al. (1993), although snow scavenging coefficients also here show the same dependence on aerosol sizes and precipitation intensity as washout coefficients, they are generally *higher* by a factor of about 5. This demonstrates the complexity of the issue, and illustrates the considerable measurement uncertainties, e.g., due to phoretic effects, and possibly due to snowflake size (as incorporated in the models of Feng, 2009, who however does not have a reasonable solution for the precipitation rate dependence). Maryon & Ryall (1996) suggest a snow scavenging coefficient that does not depend on the aerosol radius ( $\Lambda_s [s^{-1}] = 8.0 \cdot 10^{-5} q^{0.305}$ , where  $q$  is again the rain rate in mm/h), but this is in very poor agreement with measurements, and it seems reasonable to incorporate the factorial dependences from the washout coefficient as described above, and for simplicity not incorporate any scaling factors, as these would in any case not be well documented on the basis of the available literature.

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## **4. Deposition velocity data derived from Nordic measurements**

An extra effort was made to search for measurement data of Nordic origin for contaminant deposition velocities to plants (and soil). Although of course such values would be generic (i.e. not country specific), it was thought to be advantageous to make use of the Nordic network that the PardNor activity comprises to look deep into any available databases that might exist in the Nordic countries.

### **4.1 Dry deposition velocity measurements in Lund, Sweden**

A large precipitation collector, consisting of two  $1 \times 2 \text{ m}^2$  aluminium sheets slightly angled with respect to the horizontal, was used to collect samples of both dry and wet deposition. The dry deposition was measured by wiping the surface of the aluminium sheets with paper towels moistened with an organic solvent. The precipitation collector was wiped daily during the whole dry deposition period (until May 7). A centrifugal pump was used to obtain air samples for the determination of the activity of ground level air. The air was drawn through  $0.50 \times 0.50 \text{ m}^2$  glass-fibre filters mounted vertically above the ground. During the first period of deposition, the night between the 27 and 28 April, there are unfortunately no air concentration measured in Lund. Therefore the data from measurements at a sampling station at Ljungbyhed was used.

The calculated deposition velocities for the aluminium surface are shown in Table 4.1 and in Figure 4.1. Three sources of systematic errors may influence the calculation of the dry deposition velocities; the duration of the deposition took place, the air concentration and the fact that not all the material was wiped off the aluminium sheets. The fact that perhaps not all the deposited material was collected may decrease the velocities by about 10%. The high dry deposition velocities obtained on May 2, 3 and 4 could be an effect of an influence of wet deposition due to a morning mist which had not lifted before the measurements were made. It should be noted that the deposition velocity investigations were not made to crop surfaces, which would be expected to have higher deposition velocities for the same contaminants, due to the much greater surface roughness of the crops.

### **4.2 Dry deposition velocity measurements in Studsvik, Sweden**

During the period April 28 to 30, dry deposition was measured at 4 sites in the Studsvik area, in the south-east part of central Sweden. The calculations were made using time-integrated air concentrations and sampling of grass. Corrections are made for radioactive decay during the sampling process. The use of coal filters made it possible to distinguish between particulate and gaseous form of iodine. The results are shown in Table 4.2. A figure of the same results has also been published by Devell (1991). The deposition velocities in this study are higher than those observed in most studies after the Chernobyl accident and do not follow the same trends with respect to contaminant characteristics. It is possible that this demonstrates that even the slightest

wet deposition (which is a much more powerful mode of deposition) can greatly influence the picture.

Table 4.1. Dry deposition velocities ( $\text{cm s}^{-1}$ ) at Lund during the dry period 28 April-7 May 1986.

Nuclide	28 April <sup>a</sup> 17:30	29 April <sup>b</sup> 20:30	2 May <sup>a</sup> 09:15	3 May <sup>a</sup> 11:15	4 May <sup>a</sup> 10:05	5 May <sup>a</sup> 08:40	6 May <sup>b</sup> 09:20	7 May <sup>b</sup> 09:00
<sup>95</sup> Zr	0.180 ± 0.004	-	-	1.77 ± 0.06	6.8 ± 0.3	0.37 ± 0.05	0.24 ± 0.01	0.36 ± 0.05
<sup>95</sup> Nb	0.136 ± 0.002	-	-	1.74 ± 0.05	10.4 ± 0.2	1.20 ± 0.03	0.29 ± 0.01	0.23 ± 0.02
<sup>99</sup> Mo	-	-	9.5 ± 3.5		3.6 ± 0.7	0.5 ± 0.5	0.6 ± 0.2	0.1 ± 0.1
<sup>99</sup> Tc		-					0.40 ± 0.01	0.12 ± 0.01
<sup>103</sup> Ru	0.090 ± 0.002	0.22 ± 0.03	6.7 ± 0.4	1.72 ± 0.09	3.2 ± 0.05	0.39 ± 0.01	0.31 ± 0.01	0.22 ± 0.01
<sup>106</sup> Rh	0.068 ± 0.011	-	-	-	-	-	-	-
<sup>132</sup> Te	-	-	6.2 ± 0.1	9.79 ± 0.22	1.84 ± 0.01	0.195 ± 0.002	0.336 ± 0.004	0.102 ± 0.002
<sup>131</sup> I	0.032 ± 0.003	0.029 ± 0.001	1.02 ± 0.02	3.87 ± 0.01	0.0461 ± 0.0001	0.107 ± 0.002	0.074 ± 0.001	0.047 ± 0.001
<sup>134</sup> Cs	0.012 ± 0.001	-	1.6 ± 0.1	0.49 ± 0.05	0.40 ± 0.04	0.104 ± 0.004	0.32 ± 0.01	0.16 ± 0.03
<sup>136</sup> Cs	-	-	-	-	0.43 ± 0.17	0.10 ± 0.01	0.27 ± 0.03	0.13 ± 0.01
<sup>137</sup> Cs	0.015 ± 0.001	-	2.4 ± 0.1	0.45 ± 0.05	0.51 ± 0.03	0.109 ± 0.003	0.34 ± 0.01	0.18 ± 0.01
<sup>140</sup> Ba	0.113 ± 0.016	-	-	2.47 ± 0.16	4.9 ± 0.2	0.32 ± 0.02	0.30 ± 0.02	0.26 ± 0.02
<sup>140</sup> La	-	-	-	-	-	-	0.30 ± 0.01	0.28 ± 0.02
<sup>141</sup> Ce	0.19 ± 0.01	-	-	1.67 ± 0.04	7.0 ± 0.1	0.15 ± 0.02	0.193 ± 0.006	0.51 ± 0.02
<sup>144</sup> Ce	0.22 ± 0.01	-	-	1.50 ± 0.10	-	-	0.26 ± 0.03	0.009 ± 0.05
<sup>239</sup> Np	0.14 ± 0.01	-	-	-	-	-	0.08 ± 0.01	0.22 ± 0.04

<sup>a</sup>Based on air activity concentration at Ljungbyhed.

<sup>b</sup>Based on air activity concentration at Lund.

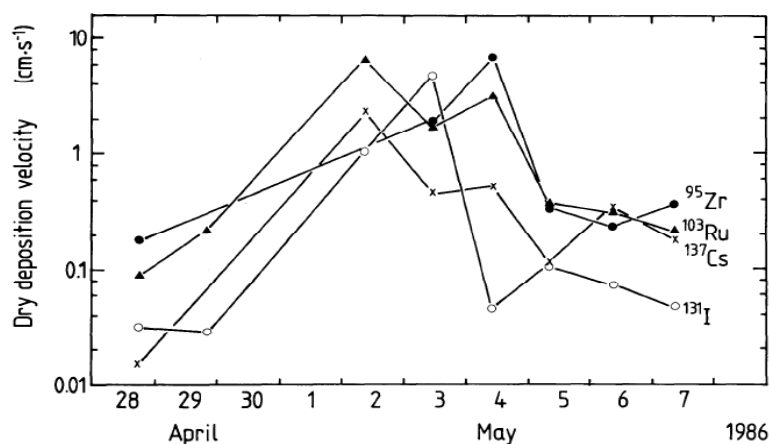


Fig. 4.1. Dry deposition velocities at Lund (aluminium surface). Published in Erlandsson & Isaksson (1988).

Table 4.2. Dry deposition velocities ( $\text{cm s}^{-1}$ ) on grass surfaces at Studsvik in the south-east part of central Sweden. Duplicate grass samples were taken at some locations (A & B).

Nuclide	Studsvik A	Studsvik B	Tranvik A	Tranvik B	Byggninge	Horsvik	Mean
$^{103}\text{Ru}$	0.58		1.0		0.44	0.33	0.59
$^{131}\text{I}^{\text{a}}$	1.7	1.9	3.7	2.7	2.0	0.99	2.3
$^{131}\text{I}^{\text{b}}$	0.54	0.56	1.1	1.1	0.62	0.30	
$^{134}\text{Cs}$	0.094		0.25		0.13	0.057	0.13
$^{137}\text{Cs}$	0.17	0.11	0.22	0.23	0.11	0.055	0.15
$^{141}\text{Ce}$	0.69		4.8			0.98	0.22
$^{239}\text{Np}$	0.54		3.2		0.29		0.13

<sup>a</sup>Iodine in particulate form.

<sup>b</sup>Correction made accounting for 70 % of iodine in gaseous form.

### 4.3 Dry deposition velocity measurements in Roskilde, Denmark

Measurements were made at Roskilde, Denmark during the passage of the first contaminated cloud from the Chernobyl accident, i.e. at midday on the 27<sup>th</sup> of April 1986. Deposition took place at a wind speed of 3 m/s, and at a height of 8 m above the ground. The atmospheric stability category was estimated to be B-C. Measurements were made of deposition velocity to a variety of different surfaces typically encountered in an inhabited area (Roed, 1990). Table 4.3 shows the deposition velocities of different contaminants to short grass cover, which may be representative of some types of agricultural crops. The figures shown for iodine are for the elemental gaseous form, whereas the data for other contaminants are for aerosols. In-line with the observations made elsewhere at this distance from the Chernobyl NPP of contaminant particle sizes (see Chapter 2), comparatively low deposition velocities were found for the volatile radionuclide aerosols (here represented by caesium and ruthenium).

Table 4.3. Dry deposition velocities of Chernobyl contaminants to short grass measured in Roskilde, Denmark on the 27<sup>th</sup> of April 1986. Physicochemical forms of the deposited contaminants are also indicated: elemental iodine gas, aerosols belonging to the ‘volatile’ group having formed small particles (typically <1 µm), and aerosols belonging to the ‘refractory’ group having formed somewhat larger particles (typically ca. 4 µm).

Contaminant	Iodine	Caesium	Ruthenium	Barium	Cerium	Zirconium
Physicochem. form	Elemental I <sub>2</sub> gas	Volatile aerosol	Volatile aerosol	Refract. aerosol	Refract. aerosol	Refract. aerosol
V <sub>d</sub> (10 <sup>-4</sup> m/s)	22	4.3	4.1	5.8	7.7	7.1

As mentioned in Chapter 2, surface roughness is very important in relation to deposition velocity. Table 4.4 shows the measurement data that was mentioned in Chapter 2 on the influence of the length of grass (Roed, 1990). The deposition velocities to different grassed areas in Roskilde, Denmark, are seen to vary considerably, as the grass had very different length in some places. However, if the length of the grass is taken into account (by dividing with the grass mass per unit area), the results are consistent within 10 %.

Table 4.4. Deposition velocities ( $v_d$ , in units of 10<sup>-4</sup> m/s) and bulk deposition velocities ( $B_d$ : deposition velocities divided by grass mass per unit area, in units of 10<sup>-4</sup> m<sup>3</sup> s<sup>-1</sup> kg<sup>-1</sup>) measured on areas with different grass lengths in Roskilde, Denmark.

Sample no.	Parameter	Cs-137	Cs-134	I-131
1384	$v_d$	4.3	4.4	22
1384	$B_d$	21	21	110
1387	$v_d$	1.8	1.5	18
1387	$B_d$	10	8.7	100
1388	$v_d$	8.8	7.2	93
1388	$B_d$	10	8.5	110
1391	$v_d$	6.0	6.6	86
1391	$B_d$	7.9	8.7	110
1392	$v_d$	7.4	9.9	120
1392	$B_d$	9.1	12.0	140

#### 4.4 Dry deposition velocity measurements in Konala and Nurmijärvi, Finland

Deposition velocities were also measured to bare soil at Konala and Nurmijärvi in Finland for a range of radiocontaminants arising from the Chernobyl accident. Table 4.5 shows the recorded values (Kostiainen, 2010). The values are however here much higher than those generally recorded for dry deposition of these contaminants, so it seems evident that much of the deposition must here be wet, unless a fraction of the particles at this location might for some reason be much larger than those recorded elsewhere in Europe at similar distances from the Chernobyl NPP (e.g., Reineking et al., 1987; Rulik et al., 1989; Dorrian, 1997).



Table 4.5. Derived deposition velocities (bare soil) for particles at two local areas of Finland after the Chernobyl reactor accident.

Nuclide	Time-integrated activity concentrations in air for a period of 24 hours (29.-30.4.1986)		Measured total depositions for the same 24 h period (STUK-B-VALO 44) (29.-30.4.1986)		Deposition velocities ( $v_d$ ) of particles calculated from previous values in this table	
	(Bq·s/m <sup>3</sup> )		(Bq/m <sup>2</sup> )		(m/s)	
	Konala	Nurmijärvi	Konala	Nurmijärvi	Konala	Nurmijärvi
Ru-103	14947	6540	570	150	0.0381	0.0229
I-131	1282463	595800	3040	670	0.0024	0.0011
I-133	108240	49457	240	350	0.0022	0.0071
Ba-140	16847	9000	500	560	0.0297	0.062
Cs-134	13263	6513	440	720	0.0332	0.11
Cs-137	22719	10776	770	870	0.0339	0.0807

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## 5. Contaminant weathering from crops

In ECOSYS, the process of natural weathering<sup>1</sup> is for all radionuclides/contaminants and all crops assumed to take place with a half-life of 25 days. This half-life estimate does not include the decrease in contaminant concentration due to growth dilution<sup>2</sup>, which is modelled elsewhere in ECOSYS. Various workers have over the years estimated the weathering half-life according to experimental findings. The majority of experiments were carried out prior to the Chernobyl accident, using tracers that more or less successfully reflected the contaminants that were believed to be released in a large nuclear power plant accident (particles in some experiments were much too large to be relevant for widespread atmospheric dispersion) (Martin, 1964; Cline et al., 1965; Milbourn & Taylor, 1965; Heinemann & Vogt, 1980; Kirchmann et al., 1966; Chadwick & Chamberlain, 1970; Krieger & Burmann, 1969, Mück et al., 1994).

### 5.1. General data collation

A limited data collation was performed in order to derive updated statistical information on weathering rates. The main sources of information were: The comprehensive compilation by Miller & Hoffmann (1983) – covering most of the pre-Chernobyl data mentioned above; Tecdocs from the International atomic energy agency: IAEA (1996) and the recently published IAEA (2010); and finally data from the BIOMOVs-project (1991) covering post-Chernobyl caesium and iodine. Nordic data are to some extent covered in the publications above e.g. weathering rates from sites in Sweden (Tranvik), Finland (Loviisa) and Denmark (Roskilde) are available for the post-Chernobyl period through BIOMOVs (1991). An effort was also put on getting additional Nordic data from PARDNOR project partners, but very limited amounts of information were discovered. Combined means and overall standard deviations for the compiled data were derived using a similar approach as described in the publication by Hosseini et al. (2008).

Since growth dilution is treated separately from weathering rates in ECOSYS, the data used for generating statistical information should preferably be per unit area (excluding the effect of growth dilution). Unfortunately, it is not always clear from the various experimental descriptions whether the derived factors include growth dilution or not. For the sake of simplicity, no attempt was made here to separate between weathering rates derived per unit mass (including growth dilution) and per unit area. This will, of course, have an impact on the statistical estimates, e.g. the calculated mean is expected to be lower than the actual value. In Miller & Hoffmann (1983) the geometric means (with ranges) for all plants and elements were 8.6 (2.8-19) and 11 (4.5-34) days – derived per unit mass and per unit area, respectively.

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<sup>1</sup> Loss of contamination from plants due to wind, rain and fog etc

<sup>2</sup> Decrease in radioelement concentration due to biomass increase during plant growth. The growth dilution rate strongly depends on the growth stage at the time of contamination as illustrated by e.g. IAEA (2010).

The results from our data collation are summarised in Table 1. Most data pertain to grass, and this plant category is therefore presented separately. The other category ('All') covers a broad group of crops (including grass, cereals, leafy vegetables, fruit, and root vegetables). Accordingly, radioelements have been divided in 5 categories (see Table 1). Here the 'All' category covers the 16 elements included in the database (Be, Ce, Co, Cr, Cs, Fe, Hg, I, Mn, Pb, Pu, Ru, Sr, Tc, Zn, Zr). Most data, however, concern iodine, caesium and strontium, thus separate weathering rates have been calculated for these three elements. No separation was made between elemental and particular iodine, since the data does not indicate any major differences in weathering rates. Furthermore, no clear differences were observed between caesium, strontium and the 'all' elements category, whereas weathering rates are lower for iodine. Consequently, the element category 'all except iodine' is also included in Table 5.1.

Table 5.1. Weathering rates (days) for different radioelement-plant combinations

Plant group	Elements	Mean	SD	n	Min	Max
All	All	18.8	12.4	208	2.4	54
All	All except iodine	23.2	12.3	138	3.1	54
All	Caesium	21.0	12.1	39	3.1	49
All	Strontium	21.8	10.8	20	9.0	47
All	Iodine	10.3	6.7	70	2.4	45
Grass	All	11.1	5.0	94	3.1	29
Grass	All except iodine	13.9	5.4	37	3.1	29
Grass	Caesium	12.7	4.6	19	3.1	27
Grass	Strontium	12.8	4.1	5	9.0	19
Grass	Iodine	9.3	3.6	57	3.4	24

Some studies have reported that weathering half-times may follow functions of two (or even more) exponentials (e.g. Ertel et al., 1989; BIOMOVs, 1991). Such data are not considered here.

It needs to be pointed out that ECOSYS make use of weathering rates only for crop types where the whole plant is used (i.e. for maize silage, beet leaves, leafy vegetables, and grass). For plants where only specific parts are used for animal feed or human consumption (i.e. Corn cobs, beet, potatoes, cereals, fruit, root vegetables, fruit vegetables, and berries) translocation factors are used instead of weathering rates. Thus adjusting the default weathering rates will not influence the radionuclide concentrations in these crops. Review of translocation factors is not part of the work tasks of PARDNOR.

The weathering rate variability, as represented by the ranges (and standard deviations) in Table 5.1, may be attributable to factors such as the physicochemical forms of the contaminants and unaccounted growth dilution (as discussed above), but another important influence could well be that the weathering processes (particularly the rain events) were of different nature in the different experiments (not always clearly described). This would be consistent with the finding of Wilkins (1987) that natural removal of Chernobyl radiocaesium from roof tiles appeared to occur only during periods of rainfall. Also, Kinnersley et al. (1996) found that when 2-8  $\mu\text{m}$  tracer labelled silica particles had deposited to wheat and bean, the effect of natural weathering on plant contaminant concentrations was in dry conditions negligible compared to the effect of growth dilution, whereas simulated rainfall seemed to have

a marked effect on the contaminant concentrations. It should be stressed that label leaching from the silica particles can not entirely be ruled out in that experiment. Also, the amounts of water applied in the experiment were very high compared with any natural scenario.

## 5.2. Influence of rain on weathering of contaminants on crops

Although half-lives may seem fairly consistent in magnitude, they all relate to specific weather conditions (generally not much rain), and the weathering half-life could well be considerably longer during a dry period than during one with prolonged and heavy rain. For instance, Madoz-Escande et al. (2004) showed that for bean, the first two rainfall incidents caused major decreases in leaf contamination, whereas later rainfall had much less influence. Inspired by this, work was carried out by Madoz-Escande et al. (2005) to examine the weathering process of various radionuclides on grass, under influence of rainfall with different characteristics (intensity, frequency and length of time period between contamination and first rainfall). The aerosols, which were produced by heating a multi-radionuclide source to 2800 °C, were found to have an average aerodynamic diameter of 3.5 µm. Caesium was found to be highly soluble from these particles (ca. 80 % dissolved in 5 minutes), whereas tellurium dissolution was weak (only 8 % after 2 hours). The caesium tracer could thus be applied in an estimation of the effect of rain showers on highly soluble radiocaesium, as was recorded after the Chernobyl accident, whereas the low-solubility particulate tellurium tracer could perhaps be used to mimic the ‘refractory’ particles that were widely dispersed by the Chernobyl accident and had a similar size distribution. Madoz-Escande et al. (2005) carried out four experiments with different rain intensities and frequencies (but same total rain amount in all cases), and found that the resultant weathering could be described by an equation of the form  $A(t) = A_1 \exp(-\lambda t) + A_2$ . Weathering was determined as Bq/m<sup>2</sup> on grass at the given time divided by Bq/m<sup>2</sup> initial contamination on the grass (rather than applying specific activities), and thus does not include growth dilution effects. Table 5.2 shows the recorded factors in the formula in relation to each of the four examined weathering conditions.

Table 5.2. Experimentally derived factors for the above formula for weathering on grass, for different rainfall conditions. Values for high-solubility <sup>137</sup>Cs and low-solubility <sup>123m</sup>Te from 3.5µm (average aerodynamic diameter) particles. Data from Madoz-Escande et al. (2005).

Rainfalls	Radionuclide	A <sub>1</sub>	A <sub>2</sub>	λ [days <sup>-1</sup> ]	T <sub>½</sub> [days]
Twice a week 8 mm/h*	<sup>137</sup> Cs	0.86	0.14	0.12	5.7
	<sup>123m</sup> Te	0.99	0.01	0.06	12.0
Once a week 8 mm/h*	<sup>137</sup> Cs	0.87	0.13	0.19	3.6
	<sup>123m</sup> Te	0.91	0.09	0.11	6.2
Twice a week 30 mm/h*	<sup>137</sup> Cs	0.91	0.09	0.06	11.7
	<sup>123m</sup> Te	1.00	0.00	0.04	19.7
Once a week 30 mm/h*	<sup>137</sup> Cs	0.88	0.12	0.07	9.9
	<sup>123m</sup> Te	1.00	0.00	0.05	12.7

\* Total rainfall was in all cases 14 mm/week; In those cases where the rain was applied over two rainfalls per week, the first rainfall occurred only 2 days after the contamination, but when the rain was applied in only one rainfall per week, the first rainfall occurred 6 days after the contamination.

As can be seen from Table 5.2, weathering half-lives for  $^{137}\text{Cs}$  are generally considerably shorter than those for the less soluble  $^{123\text{m}}\text{Te}$ , which to a greater extent remains in particle form. However, had the particles been very large, they would most likely have been removed rapidly by rain (Roed & Andersson, 1996). It is also seen that the duration of the rainfall event is important: if the 14 mm/week are applied in one rainfall (on day 6), this is more efficient in washing the contamination off than when the same amount of rain was applied over two rainfall events, where the first started already on day 2. Finally, also the rain intensity is seen to have an effect. Here a long, but not very intensive rain shower seems to be more efficient in removing the contamination than a short but powerful one. This is consistent with results of spraying water to remove Chernobyl radiocaesium from a contaminated wall: also here the duration of spraying was more important than the intensity (Andersson, 1991).

Table 5.3 shows a set of simple single-exponential function half-lives found for a similar rain scenario. The very low values for bean and wheat may possibly reflect that the first rainfall occurred very early (after only one day). The values for radish and lettuce are in better agreement with the above values for grass. For comparison, Krieger & Burmann examined the weathering of strontium and caesium applied on grass as readily soluble chlorides. In areas protected against rain, the weathering half-life (including growth dilution) was found to be 15-18 days, whereas it was in areas subjected to natural rain (in the early stage less than 2 mm/day; first rain after 3 days) typically found to be only 3-7 days.

Table 5.3. Experimentally derived simple exponential function weathering half-lives, for contaminants on a range of crops and a specific set of rain conditions. Values for high-solubility  $^{137}\text{Cs}$  and  $^{85}\text{Sr}$  and low-solubility  $^{123\text{m}}\text{Te}$  from  $3.5\mu\text{m}$  (average aerodynamic diameter) particles. Data from Madoz-Escande & Santucci (2005).

Rainfalls	Radionuclide	$\lambda$ [ $\text{d}^{-1}$ ]	$T_{1/2}$ [d]	Time rain 1 [d] <sup>#</sup>
Bean: Twice a week, 7-10 mm/h*	$^{137}\text{Cs}$	0.4	1.7	1
	$^{85}\text{Sr}$	0.45	1.5	1
Wheat: Twice a week, 7-10 mm/h*	$^{137}\text{Cs}$	0.8	0.9	1
	$^{85}\text{Sr}$	1.2	0.6	1
Radish: Twice a week, 7-10 mm/h*	$^{137}\text{Cs}$	0.33	2.1	2
	$^{123\text{m}}\text{Te}$	0.06	11.6	2
Lettuce: Twice a week, 7-10 mm/h*	$^{137}\text{Cs}$	0.19	3.6	2
	$^{123\text{m}}\text{Te}$	0.05	13.9	2

\* Total rainfall was in all cases 16-20 mm/week. <sup>#</sup> Time between contamination and first rainfall.

Although Tables 5.2 and 5.3 are informative in qualitatively increasing the understanding of the mechanisms governing the natural weathering process of radioactive contaminants on vegetation, data for more crops and for different total amounts of rain per week would be required to have a really firm foundation for modelling the weathering process in ARGOS and RODOS on the background of actual or predicted rain data. Nevertheless, the half-lives in Table 5.3 show that for realistic scenarios, the weathering half-life can be much shorter than the 25 days assumed as default in ECOSYS, and it may be recommended to adjust the weathering half-life somewhat in ARGOS and RODOS, according to whether the weather is dry

or rainy over long periods, and also according to the physicochemical form or solubility of the contaminant. Indeed, the ECOSYS weathering half-life of 25 days seems to reflect dry conditions: together with the ECOSYS typical default grass growth dilution half-life of 20 days, this gives a total half-life of about 16 days corresponding to the 15-18 days in Krieger & Burmann's rain protected scenario.

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## 6. Crop weathering data derived from Nordic measurements

An extra effort was also made to search for measurement data of Nordic origin for crop weathering data (half-lives of the natural weathering process reducing the level of contamination on the surface of crops after deposition). This parameter would depend on the weather type (particularly the rain events, as described in Chapter 5) over the first weeks after the contaminants deposited. Therefore, some local variation could be expected. It was thought to be advantageous to make use of the Nordic network that the PardNor activity comprises to look deep into any available databases that might exist in the Nordic countries. However, very little Nordic measurement data was found for this parameter, which is summarised in Table 2.2 (BIOMOVS, 1991). The very low values for the Loviisa location may possibly indicate effects of growth dilution.

Table 6.1. Weathering half-lives from pasture vegetation for I-131 and Cs-137 (days), calculated on the basis of Chernobyl contamination measurements.

Location	I-131	Cs-137
Loviisa, Finland	3.5	3.1
Tranvik, Sweden	6.6	15
Roskilde, Denmark	13	9.0

### Reference

BIOMOVS (1991). Scenario 4. Multiple Model Testing using Chernobyl fallout data of I-131 and Cs-137 in forage, milk, beef and grain. H. Kohler, S.-R. Peterson & F.O. Hoffmann (eds.). Technical Report 13, vols. I and II, Stockholm: National Institute of Radiation Protection.



## 7. ECOSYS runs to demonstrate the effect of deposition velocity and weathering parameter improvement

### 7.1. Introduction

The ECOSYS model is a radiological model for assessing contamination of foodstuffs and radiation exposure after a release of radionuclides. It is essentially the food chain module in the European state-of-the-art decision support systems RODOS and ARGOS.

The ECOSYS model was developed in the 1980's, and it is based on environmental conditions of Southern Germany. It is therefore necessary to adjust some parameters in the model when it is used for other environments, and when better knowledge becomes available for e.g. physical parameters in the model. This study has focus on the effect of changing the deposition velocities and the weathering half life in the Excel version of ECOSYS.

### 7.2. Material and methods

The products in the study include grass (intensive and extensive), leafy vegetables, fruit vegetables and cow milk. The considered isotopes are  $^{137}\text{Cs}$ ,  $^{90}\text{Sr}$  and  $^{131}\text{I}$ , with the default fractions for iodine, i.e. 1/3 aerosol bound, 1/3 elemental and 1/3 organic bound.

The deposition in all scenarios is specified as dry deposition on the 1<sup>st</sup> of August, with the time-integrated activity concentration in air equal to 200 Bq h/m<sup>3</sup>.

#### *Deposition velocities in ECOSYS*

The deposition velocities,  $V_d$ , in ECOSYS refer to fully developed plant canopies. They are, though, modified in proportion to the ratio of the actual and maximum leaf area index, LAI, if the actual LAI is less than the maximum. The dry deposition of radionuclides to crops is calculated from the deposition velocity and the time-integrated activity concentration in air.

For all crops except grass and fruits, ECOSYS uses  $V_d=2.0$  mm/s as the default aerosol deposition velocity. It uses  $V_d=20$  mm/s and  $V_d=0.2$  mm/s for, respectively, elemental iodine and organic iodine gas. For grass, the default for aerosols is  $V_d=1.5$  mm/s, while it is 15 mm/s and 0.15 mm/s for elemental and organic iodine, respectively. In lack of better information, the default  $V_d$  to fruits is 2.5 times higher than to other crops.

#### *Adjusting deposition velocities*

The findings from a literature study on measured dry deposition velocities of aerosols and iodine to agricultural crops is summarised in Table 7.1.

Table 7.1. Comparison between ECOSYS default deposition velocities,  $V_d$ , to agricultural crops and a new dataset derived from a literature study (see Chapter 2 for details on parameter derivation).

Dataset	$V_d$ (1 $\mu\text{m}$ aerosol), mm/s	$V_d$ (4 $\mu\text{m}$ aerosol), mm/s	$V_d$ (20 $\mu\text{m}$ aerosol), mm/s	$V_d$ (elemental iodine gas), mm/s	$V_d$ (organic iodine gas), mm/s
New dataset	0.3	1.5	35	5	0.02
ECOSYS defaults	2	2	-	20	0.2

The default size of aerosol contaminants in ECOSYS is claimed to be less than 1  $\mu\text{m}$ .

In connection with the Chernobyl accident, two different aerosol size groups were observed (see Chapter 2 of this report). On this basis, the ECOSYS defaults should be appropriate for e.g. caesium. For e.g. strontium, the effect of gravitational settling should result in a considerably higher deposition velocity as compared to caesium.

The ECOSYS default deposition velocity to bare soil is 0.5 mm/s for aerosols, 3 mm/s for elemental iodine, and 0.05 mm/s for organic bound iodine. However, on the basis of the literature, it is suggested to use a value of 0.1 mm/s for aerosols of the 1  $\mu\text{m}$  range (e.g. caesium), 0.4 mm/s for aerosols of the 4  $\mu\text{m}$  range (e.g. strontium), 3 mm/s for elemental iodine, and 0.02 mm/s for organic bound iodine (see Chapter 2).

The deposition velocities for caesium and strontium in Table 7.2 have been evaluated according to the above comments and the new dataset in Table 7.1. Tables 7.2 and 7.3 summarize the considered deposition velocities. In the adjusted datasets, the particle deposition velocities for grass, fruit and lawn are evaluated in the same ratio to the values for other crops as in the default case.

Table 7.2. Default and adjusted deposition velocities (mm/s) for  $^{137}\text{Cs}$  and  $^{90}\text{Sr}$  aerosols in the ECOSYS scenarios.

Plant	$^{137}\text{Cs}$		$^{90}\text{Sr}$	
	ECOSYS defaults	Adjusted velocities	ECOSYS defaults	Adjusted velocities
	Particle	Particle	Particle	Particle
Grass Intensive	1.5	0.225	1.5	1.125
Grass Extensive	1.5	0.225	1.5	1.125
Maize	2	0.3	2	1.5
Corn cobs	2	0.3	2	1.5
Potatoes	2	0.3	2	1.5
Beet	2	0.3	2	1.5
Beet leaves	2	0.3	2	1.5
Winter barley	2	0.3	2	1.5
Spring barley	2	0.3	2	1.5
Winter wheat	2	0.3	2	1.5
Spring wheat	2	0.3	2	1.5
Rye	2	0.3	2	1.5
Oats	2	0.3	2	1.5

Leafy vegetab.	2	0.3	2	1.5
Root vegetables	2	0.3	2	1.5
Fruit vegetables	2	0.3	2	1.5
Fruit	5	0.75	5	3.75
Berries	2	0.3	2	1.5
Lawn	1.5	0.225	1.5	1.125
Soil	0.5	0.1	0.5	0.4

Table 7.3. Default and adjusted deposition velocities (mm/s) for Iodine in the ECOSYS scenarios.

Plant	ECOSYS defaults for <sup>131</sup> I			Adjusted velocities for <sup>131</sup> I		
	Particle	elemental	organic	Particle	elemental	organic
Grass Int.	1.5	15	0.15	0.225	5	0.02
Grass Ext.	1.5	15	0.15	0.225	5	0.02
Maize	2	20	0.2	0.3	5	0.02
Corn cobs	2	20	0.2	0.3	5	0.02
Potatoes	2	20	0.2	0.3	5	0.02
Beet	2	20	0.2	0.3	5	0.02
Beet leaves	2	20	0.2	0.3	5	0.02
Winter barley	2	20	0.2	0.3	5	0.02
Spring barley	2	20	0.2	0.3	5	0.02
Winter wheat	2	20	0.2	0.3	5	0.02
Spring wheat	2	20	0.2	0.3	5	0.02
Rye	2	20	0.2	0.3	5	0.02
Oats	2	20	0.2	0.3	5	0.02
Leafy vegetables	2	20	0.2	0.3	5	0.02
Root vegetables	2	20	0.2	0.3	5	0.02
Fruit vegetables	2	20	0.2	0.3	5	0.02
Fruit	5	50	0.5	0.75	12.5	0.5
Berries	2	20	0.2	0.3	5	0.02
Lawn	1.5	15	0.15	0.225	5	0.02
Soil	0.5	3	0.05	0.1	3	0.02

#### *Weathering half life in ECOSYS*

In ECOSYS, weathering describes the loss of activity from plant due to wind, rain and fog and the loss of dead plant material. The weathering loss does not include the decrease of activity concentration due to growth dilution. The default weathering half life in ECOSYS is  $T_{1/2} = 25$  days for all contaminants on all crops.

### Adjusting weathering half lives

Table 7.4 shows an expert judgement of the weathering half life,  $T_{1/2}$ . The data are based on investigations of existing databases (see Chapter 5).

Grass		Plants, general		Plants, general,
<sup>137</sup> Cs and <sup>90</sup> Sr	<sup>131</sup> I	<sup>137</sup> Cs and <sup>90</sup> Sr	<sup>131</sup> I	All nuclides
15d (10d-25d)	10d (5d-20d)	20d (10d-35d)	10d (5d-20d)	20d (5d-50d)

The values in Table 7.4 are obviously different from the default value in ECOSYS. In this study, the data in Table 7.4 were used for grass (intensive and extensive), leafy vegetables and cow milk. The ECOSYS default values were used for all parameters except the weathering half life.

### Scenario tables in ECOSYS

Tables 7.5-7.7 show the relevant parameter tables in the Scenario worksheet of ECOSYS. Tables 7.5 and 7.6 describe when the growth begins, when the harvest begins and when the harvest ends. The growing period is the difference between the date for begin of growth and the date for begin of harvest. Table 7.7 describes the feeding of lactating cows.

Table 7.5. Scenario table from ECOSYS regarding Leaf area indices, Growing periods and Yields.

Explanation: The morphological development of plants is quantified by the time-dependence of the leaf area index (LAI), which is defined as ratio of the one-sided leaf area per unit soil area. For grass the yield (kg/m<sup>2</sup> FM) is given, since for grass a good correlation exists between the leaf area and the yield. Health Phys., 64(1993), S. 237

	Grass Intensive	Grass Extensive	Lawn
Date1	01-jan	01-jan	01-jan
Yield1	0.03	0.03	0.03
Date2	15-mar	15-mar	15-mar
Yield2	0.05	0.05	0.05
Date3	16-maj	30-jun	15-maj
Yield3	1.50	1.50	0.30
Date4	31-okt	31-okt	31-okt
Yield4	1.50	1.50	0.30
Date5	01-nov	01-nov	01-nov
Yield5	0.05	0.05	0.10
Date6	31-dec	31-dec	31-dec
Yield6	0.03	0.03	0.03
Date7	31-dec	31-dec	31-dec
Yield7	0.03	0.03	0.03

Date8	31-dec	31-dec	31-dec
Yield8	0.03	0.03	0.03
Date9	31-dec	31-dec	31-dec
Yield9	0.03	0.03	0.03
Begin of growth	15-mar	15-mar	
Begin of harvest	20-apr	20-apr	
End of harvest	10-nov	10-nov	
Yield (FM, kg/m <sup>2</sup> )	time-dependent	time-dependent	time-dependent

Table 7.6. Parameter settings in ECOSYS scenario table. Danish parameters for leafy vegetables are shown in the table for comparison.

	Leafy vegetables. ECOSYS default	Leafy vegetables. Danish with normal fertilization	Fruit vegetables ECOSYS default
Date1	01-jan	10-apr	01-jan
LAI1	5.00	0.00	0.00
Date2	31-dec	01-may	15-apr
LAI2	5.00	1.00	0.00
Date3	31-dec	01-jun	01-jul
LAI3	5.00	5.00	5.00
Date4	31-dec	15-sep	01-okt
LAI4	5.00	4.00	5.00
Date5	31-dec	31-okt	31-okt
LAI5	5.00	2.50	0.00
Date6	31-dec	15-nov	31-dec
LAI6	5.00	0.00	0.00
Date7	31-dec	31-dec	31-dec
LAI7	5.00	0.00	0.00
Date8	31-dec	31-dec	31-dec
LAI8	5.00	0.00	0.00
Date9	31-dec	31-dec	31-dec
LAI9	5.00	0.00	0.00
Begin of growth	12-mar	10-apr	15-apr
Begin of harvest	01-maj	01-jun	01-aug
End of harvest	31-okt	31-okt	15-okt
Yield (FM, kg/m <sup>2</sup> )	2.0	2.0	1.5

Table 7.7. Scenario table from ECOSYS regarding feeding (kg/day) of lactating cow.

Date	Julian day	Feed1 / Index	Feed2 / Index
		Grass, intensive	Hay, intensive
		1	2
01-jan	1	0	14
20-apr	111	0	14
10-maj	131	70	0
20-okt	294	70	0
09-nov	314	0	14
31-dec	365	0	14

### 7.3. Results and discussion

#### *ECOSYS with adjusted deposition velocities*

The default deposition velocities,  $V_d$ , in ECOSYS were adjusted according to the new datasets in Tables 7.1-7.3 in order to investigate the impact on ECOSYS endpoints. The study includes grass, leafy vegetables, fruit vegetables and cow milk. The considered isotopes were  $^{137}\text{Cs}$ ,  $^{90}\text{Sr}$  and  $^{131}\text{I}$ .

The results for  $^{137}\text{Cs}$  and  $^{90}\text{Sr}$  are presented in Figures 7.1-7.10. The figures show activity concentrations together with ratios between ECOSYS runs with adjusted and default deposition velocities (cf. Tables 7.1-7.2).

The most relevant time is shortly after the release. The simulation time was, however, set to two years in order to also look at long term effects from the changed deposition velocities. The effect from soil uptake becomes increasingly important in the long term.

The  $^{137}\text{Cs}$  and  $^{90}\text{Sr}$  activity concentrations in grass decreased when  $V_d$  was decreased from the default value. The decrease was larger for caesium than for strontium due to the larger decrease of  $V_d$  in the case of caesium (Figures 7.1-7.4). The gaps in the time series are related to the start and end of harvest (Table 7.5). During the first days after the deposition, the ratio between the results from ECOSYS runs with adjusted and default  $V_d$  was equal to the ratio between the adjusted and the default  $V_d$  values. The ratio changed subsequently due to the increased importance of soil uptake together with the fact that the ratio  $V_d(\text{soil})/V_d(\text{crops})$  was different in the adjusted and the default dataset (cf. Table 7.2). (The ratio between the ECOSYS runs was constant in time equal to the ratio between the adjusted and the default  $V_d$  values when the ratio  $V_d(\text{soil})/V_d(\text{crops})$  was the same in the adjusted and the default dataset; these the simulations are not presented in the report).

The  $^{137}\text{Cs}$  concentration in leafy vegetables and fruit vegetables are shown in Figures 7.5 and 7.7, respectively. In the first period after the deposition, the concentrations from ECOSYS runs with adjusted  $V_d$  were 85.0% lower than the corresponding results with default  $V_d$ . This corresponds to the decrease of  $V_d$  from the default of 2.0

mm/s to 0.3 mm/s. After the 21<sup>th</sup> of September, the decrease changed to 83.5%. The <sup>90</sup>Sr concentration in leafy vegetables and fruit vegetables (Figures 7.6 and 7.8) similarly decreased by 25.0% in the first period after the deposition, again corresponding to the decrease of  $V_d$  from the default of 2.0 mm/s to 1.5 mm/s. After the 21<sup>th</sup> of September, the decrease changed to 23.5%.

A problem in using ECOSYS to model the contamination of leafy vegetables and the associated doses to humans, which became apparent in connection with these model runs, is that the ECOSYS modellers here use input data from the same table (with the same headings) as for all other crops, although the modelling of this specific crop has been done in a totally different way. This means that the names of the parameters are for this crop misleading, and the different model structure for leafy vegetables is not mentioned in the user guide for ECOSYS, nor in scientific papers describing ECOSYS, nor in readily available user guide material for ARGOS and RODOS, where the ECOSYS model is implemented. This is problematic and needs to be pointed out, as customisation attempts to make the model applicable for a specific locality could well lead to strange modelling assumptions that are not visible to the modeller. The special problem with modelling of leafy vegetables is that they are continuously grown and harvested over a given period of the year, but only the first of these successive harvests is directly contaminated by air pollutants. Of course this requires a different model approach than for, e.g., grains or root vegetables, where it is generally assumed that only one 'generation' of crops are harvested each year. This means that the data entered for the parameter entitled 'end of harvest' for leafy vegetables determines the time of the year, where plants no longer *grow*. It is however assumed that by this time, the relevant field is full of fully mature leafy vegetables, which are harvested slowly (thus with a reduced consumption rate) over the winter period (thus the constant contaminant concentrations in leafy vegetables after the 21<sup>st</sup> of September in Figures 16a, 16b, 17a, 17b and 22b). This continuous harvesting of leafy vegetables over a winter period may possibly be reasonable in mild climates, such as in Southern Germany (which ECOSYS is by default parameterised for), but is clearly flawed for many Nordic areas, where frost would destroy the crops. It is problematic that customisation of ECOSYS is here not possible for Nordic areas by simply changing parameters. It further seems that the parameter 'begin of harvest' is actually used to define the end of the first harvest, and thus the growth period of each 'generation' of leafy vegetables.

The activity concentrations of <sup>137</sup>Cs and <sup>90</sup>Sr in cow milk are presented in Figures 7.9 and 7.10, respectively. During the passage of the radioactive cloud, ECOSYS includes the inhalation by the cows (5 m<sup>3</sup>/hour in ECOSYS) as a minor influence on the activity concentration in the milk. During the first days, the activity concentration in milk increases as the cows start to feed on contaminated food. It then decreases, and the pattern of the time series mainly reflects the feeding regime of the lactating cows (Table 7.5). As e.g. in Figure 7.9, where the first decrease in the activity concentration of <sup>137</sup>Cs is followed by a slight increase followed by a constant level, because the feed changes from pasture grass to hay harvested earlier the same year. Then follows a period where the cows again feed on pasture grass, and where the root uptake is the dominant source for the radionuclides in the milk. Before this latter period, the ratio between the endpoint concentrations with adjusted  $V_d$  and default  $V_d$  was equal to the ratio between the adjusted and default  $V_d$  values. The long term variation of <sup>137</sup>Cs and <sup>90</sup>Sr in cow milk also reflects the activity concentration in grass (Figures 7.1-7.4). The

comparatively high concentration relationship at the very start of the curve reflects the greater relative importance of the cow's inhalation pathway compared with the cow's ingestion pathway, when crop deposition velocities are lower.

Figures 7.11a and 7.11b show the results for  $^{131}\text{I}$  as based on the datasets in Table 7.3. As compared to a scenario with the default dataset, the activity concentration in grass and cow milk decreased by 68% in the very early phase after the deposition while the corresponding decrease for leafy vegetables and fruit vegetables was 76%. For milk, the explanation for the time variation is the same as discussed above for caesium and strontium.

As illustrated by the figures, introducing corrected, particle size specific deposition velocity data instead of ECOSYS defaults can actually change contaminant concentration estimates in food products by almost an order of magnitude, for a Chernobyl type contamination scenario (caesium, strontium and iodine contaminant forms were modelled in compliance with that was recorded after the Chernobyl release). Other conceivable scenarios (e.g., involving fires) could lead to production of much larger particles for which the importance of changing ECOSYS deposition parameters could be even greater.

*ECOSYS with adjusted weathering half lives*

Figures 7.12-7.23 show the results with ECOSYS when the weathering half life was changed from the default of  $T_{1/2} = 25$  days. All other parameters had default values.

The endpoint activity concentrations of the products decreased as the weathering half life was decreased from 25 days. The maximum decrease occurred earlier as  $T_{1/2}$  was decreased from 25 days, i.e. faster decrease in the activity concentration as  $T_{1/2}$  decrease below 25 days. The opposite was seen when  $T_{1/2}$  was increased from 25 days. Tables 7.8-7.10 show maximum and minimum concentrations relative to the concentrations found by using  $T_{1/2} = 25$  days. It should be noted that the ECOSYS output has a time resolution of 2 days in the later part of the simulated time period.

The figures clearly illustrate the importance of applying correct values for the weathering half-life. Contamination estimates in the first year may be wrong by more than one order of magnitude, if the applied weathering parameter is not appropriate (e.g., if a value for dry weather is applied, when there is actually heavy rain).

Table 7.8. Minimum and maximum of  $^{137}\text{Cs}$  activity concentration as found relative to ECOSYS results with the default weathering half life  $T_{1/2} = 25$  days.

	$^{137}\text{Cs}$ Intensive grass	$^{137}\text{Cs}$ Extensive grass	$^{137}\text{Cs}$ Leafy vegetables	$^{137}\text{Cs}$ Cow milk
Minimum; $T_{1/2} = 5$ days	0.212	0.062	0.010	0.267
Date of minimum	25 August	29 August	13 September	28 August
Maximum; $T_{1/2} = 50$ days	1.82	2.44	1.95	1.69
Date of maximum	10 November	10 November	19 September	23 October



Table 7.9. Minimum and maximum of  $^{90}\text{Sr}$  activity concentration as found relative to ECOSYS results with the default weathering half life  $T_{1/2}=25$  days.

	$^{90}\text{Sr}$ Intensive grass	$^{90}\text{Sr}$ Extensive grass	$^{90}\text{Sr}$ Leafy vegetables	$^{90}\text{Sr}$ Cow milk
Minimum; $T_{1/2}=5$ days	0.063	0.060	0.031	0.094
Date of minimum	5 September	3 September	7 September	9 September
Maximum; $T_{1/2}=50$ days	2.39	3.00	1.83	2.02
Date of maximum	10 November	10 November	17 September	23 October

Table 7.10. Minimum and maximum of  $^{131}\text{I}$  activity concentration as found relative to ECOSYS results with the default weathering half life  $T_{1/2}=25$  days.

	$^{131}\text{I}$ Intensive grass	$^{131}\text{I}$ Extensive grass	$^{131}\text{I}$ Leafy vegetables	$^{131}\text{I}$ Cow milk
Minimum; $T_{1/2}=5$ days	0.213	0.141	0.017	0.213
Date of minimum	25 August	3 September	11 September	27 August
Maximum; $T_{1/2}=50$ days	1.80	2.88	1.90	1.77
Date of maximum	10 November	10 November	19 September	21 October

#### 7.4. Summary on ECOSYS model runs

The study covered ECOSYS scenarios with a dry deposition event on the 1<sup>st</sup> of August. The considered isotopes were  $^{137}\text{Cs}$ ,  $^{90}\text{Sr}$  and  $^{131}\text{I}$ . The deposition velocity and the weathering half life for the isotopes were adjusted in accordance with findings in the literature. The selected products were grass (intensive and extensive), leafy vegetables, fruit vegetables and cow milk.

The deposition velocity,  $V_d$ , to crops was changed from 2 mm/s to 0.3 mm/s for caesium, and from 2 mm/s to 1.5 mm/s for strontium. The deposition velocities to grass and lawn were specified with the same ratio to the deposition velocity to other crops as in the ECOSYS default parameter file. The ECOSYS default deposition velocity to bare soil is 0.5 mm/s for aerosols, 3 mm/s for elemental iodine, and 0.05 mm/s for organic bound iodine. On the basis of the literature studies, the deposition velocity to soil was changed to 0.1 mm/s and 0.5 mm/s for caesium and strontium, respectively. For iodine, the deposition velocities to soil were changed to 0.1 mm/s for particle, 3 mm/s for elemental iodine, and 0.02 mm/s for organic bound iodine.

The results are presented as activity concentrations and as the ratio between activities found by using the adjusted and the default parameters in ECOSYS. The ratios in the

scenarios with different deposition velocities were equal to the ratio between the adjusted and the default deposition velocity in the early phase after the deposition. The ratios increased slightly in the long term as root uptake becomes dominating, and as the ratio between the deposition velocities to soil and to crops was different in the adjusted and the default parameter dataset. The ratio  $V_d(\text{soil})/V_d(\text{crop})$  for e.g.  $^{137}\text{Cs}$  was 0.1/0.3~ 0.33 in the adjusted dataset and 0.5/2.0= 0.25 in the default dataset.

The weathering half life in ECOSYS is  $T_{1/2}= 25$  days for all crops and all isotopes. In the literature and existing databases, however, weathering half lives are generally found to be between 5 days and 50 days.  $T_{1/2}$  was changed accordingly in ECOSYS in order to study the effect on the endpoint concentrations during the first months after the deposition. The resulting activity concentrations are presented together with the ratios to the corresponding concentrations when  $T_{1/2}= 25$  days. The minimum and maximum ratios were found when  $T_{1/2}= 5$  and  $T_{1/2}= 50$ , respectively, as the activity concentration decreases faster as  $T_{1/2}$  decreases. The minimum ratios were between 0.01 and 0.27, while the maximum ratios were between 1.7 and 3.0.

The ECOSYS runs in this chapter clearly demonstrated the importance of applying correct values for the deposition velocities and weathering rates of contaminants on crops, as order of magnitude changes in crop contaminant concentrations were in some cases recorded where ECOSYS default values of these parameters were changed to more realistic ones.

Figure 7.1. Cs-137 concentration in intensive grass. Results with default and adjusted deposition velocities Vd. The secondary y-axis shows the ratio between results with adjusted and default deposition velocities.

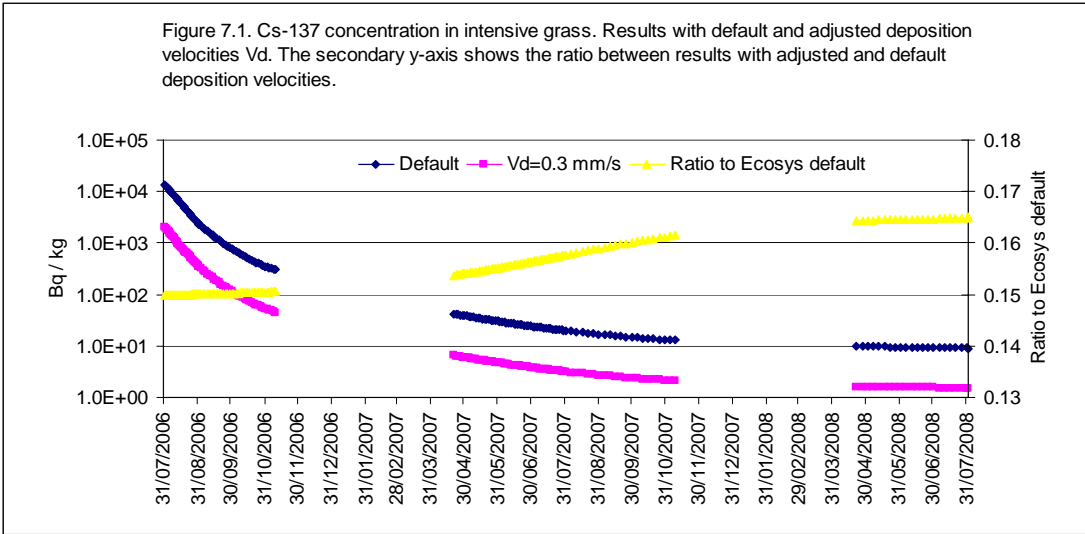


Figure 7.2. Sr-90 concentration in intensive grass. Results with default and adjusted deposition velocities Vd. The secondary y-axis shows the ratio between results with adjusted and default deposition velocities.

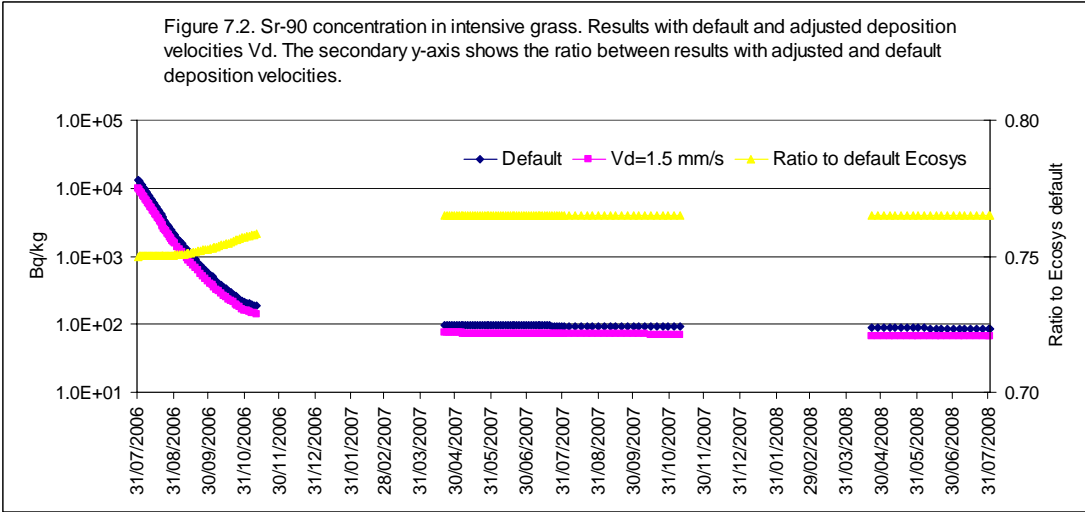


Figure 7.3. Cs-137 concentration in extensive grass. Results with default and adjusted deposition velocities Vd. The secondary y-axis shows the ratio between results with adjusted and default deposition velocities.

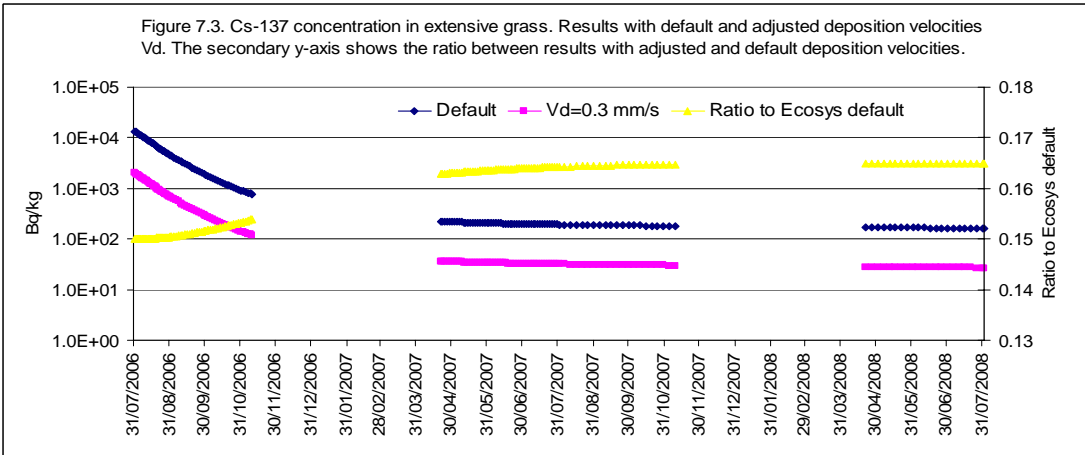


Figure 7.4. Sr-90 concentration in extensive grass. Results with default and adjusted deposition velocities Vd. The secondary y-axis shows the ratio between results with adjusted and default deposition velocities.

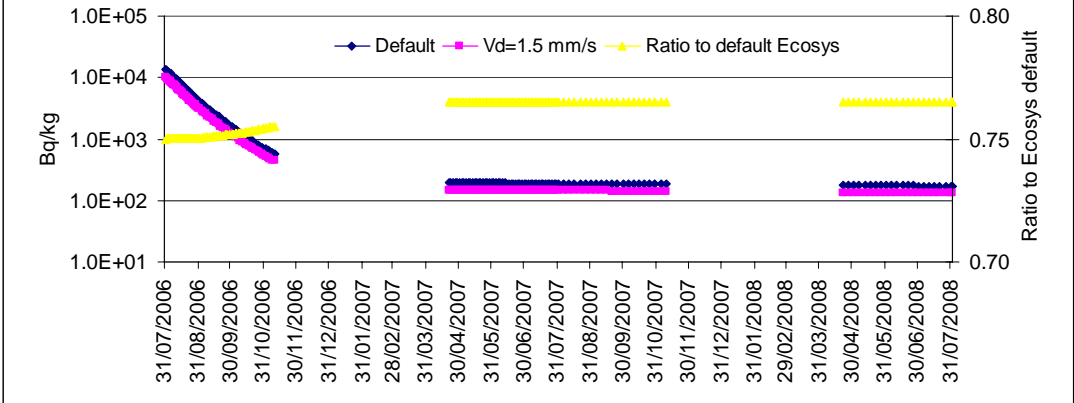


Figure 7.5. Cs-137 concentration in leafy vegetables. Results with default and adjusted deposition velocities Vd. The secondary y-axis shows the ratio between results with adjusted and default deposition velocities.

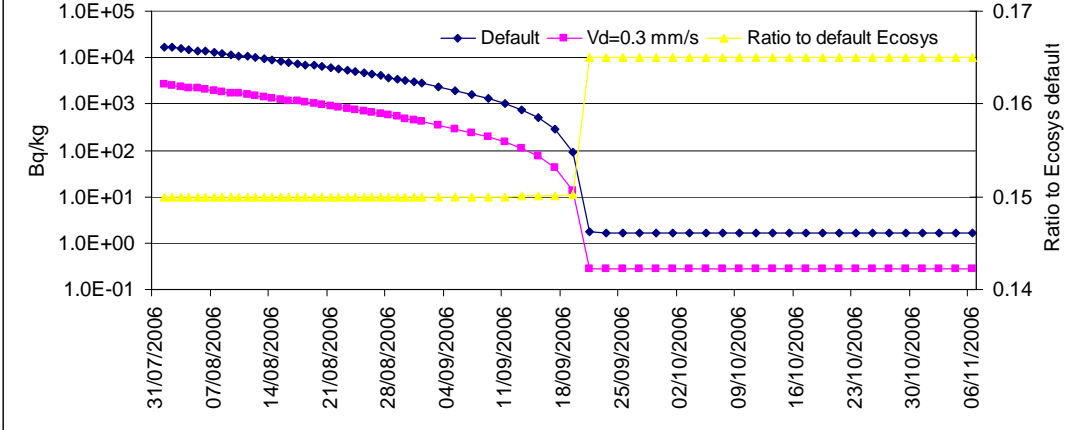


Figure 7.6. Sr-90 concentration in leafy vegetables. Results with default and adjusted deposition velocities Vd. The secondary y-axis shows the ratio between results with adjusted and default deposition velocities.

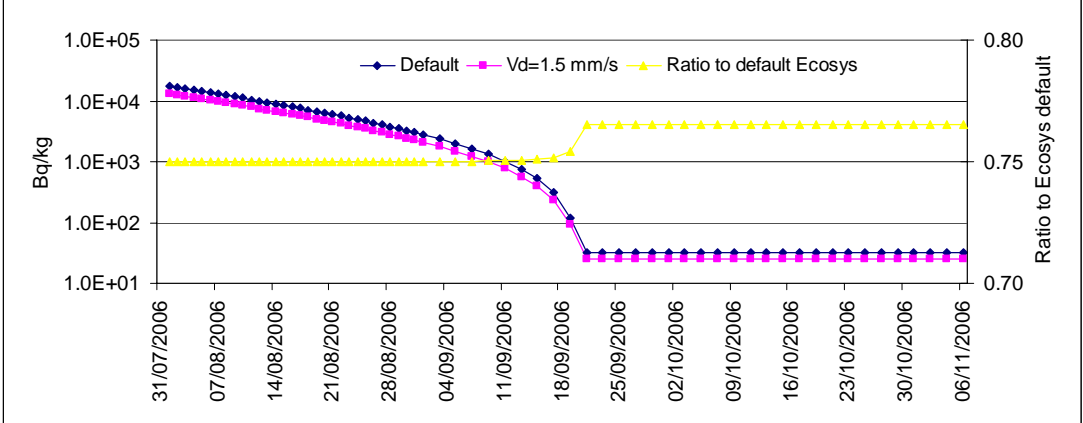


Figure 7.7. Cs-137 concentration in fruit vegetables. Results with default and adjusted deposition velocities Vd. The secondary y-axis shows the ratio between results with adjusted and default deposition velocities.

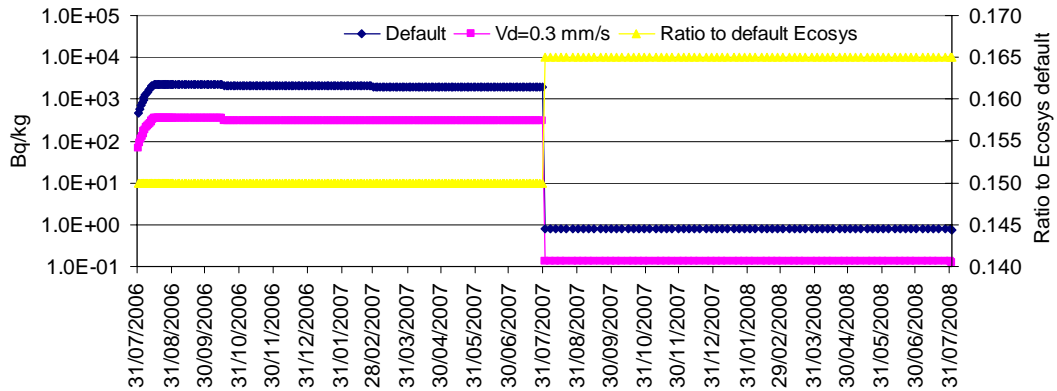


Figure 7.8. Sr-90 concentration in fruit vegetables. Results with default and adjusted deposition velocities Vd. The secondary y-axis shows the ratio between results with adjusted and default deposition velocities.

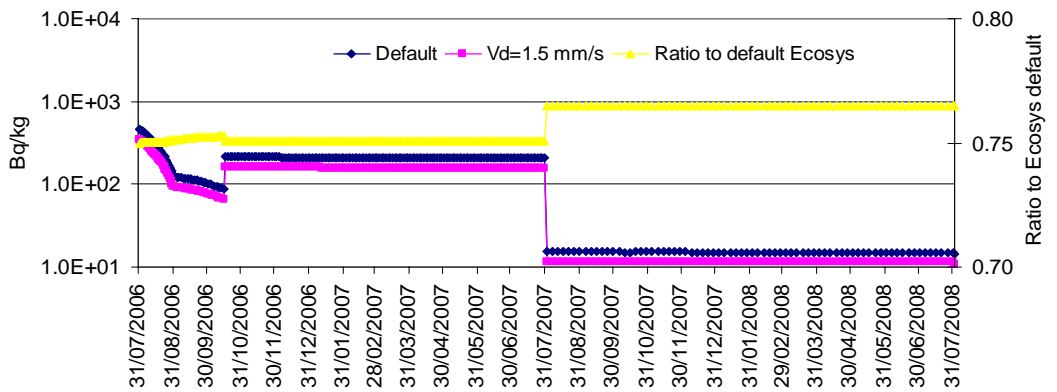


Figure 7.9. Cs-137 concentration in cow milk. Results with default and adjusted deposition velocities Vd. The secondary y-axis shows the ratio between results with adjusted and default deposition velocities.

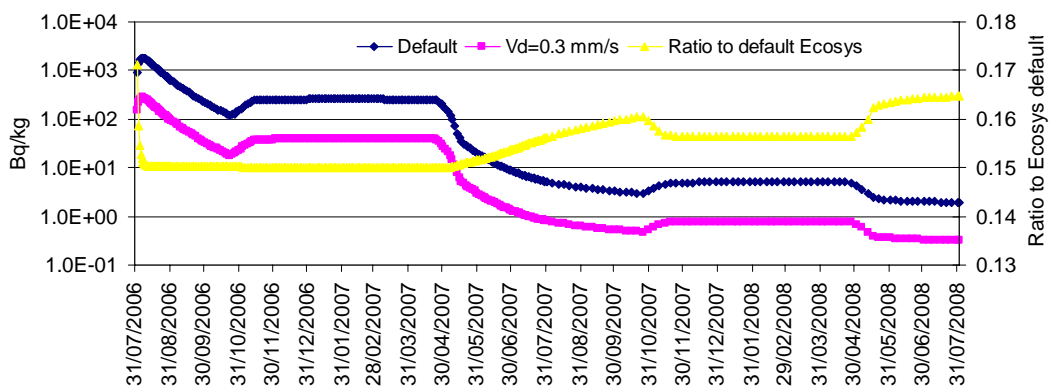


Figure 7.10. Sr-90 concentration in cow milk. Results with default and adjusted deposition velocities Vd. The secondary y-axis shows the ratio between results with adjusted and default deposition velocities.

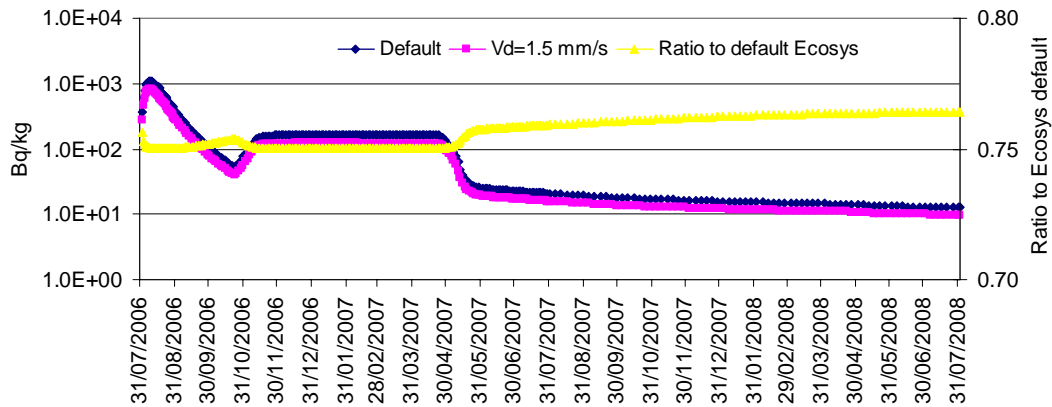


Figure 7.11a. I-131 concentration in selected products with standard and adjusted (see text) deposition velocities. The secondary y-axis shows the ratio between results with adjusted and default deposition velocities.

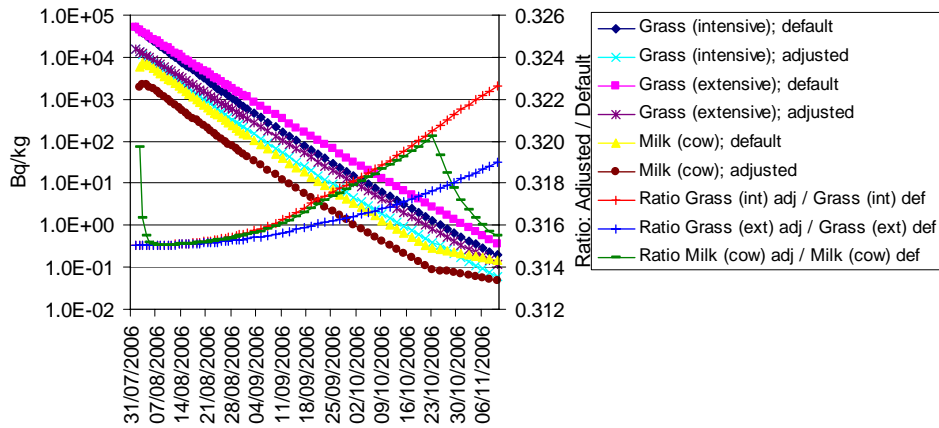
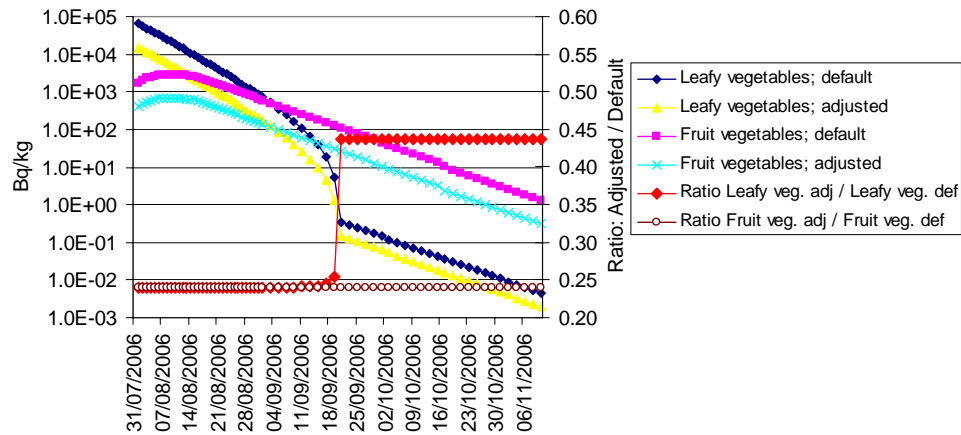
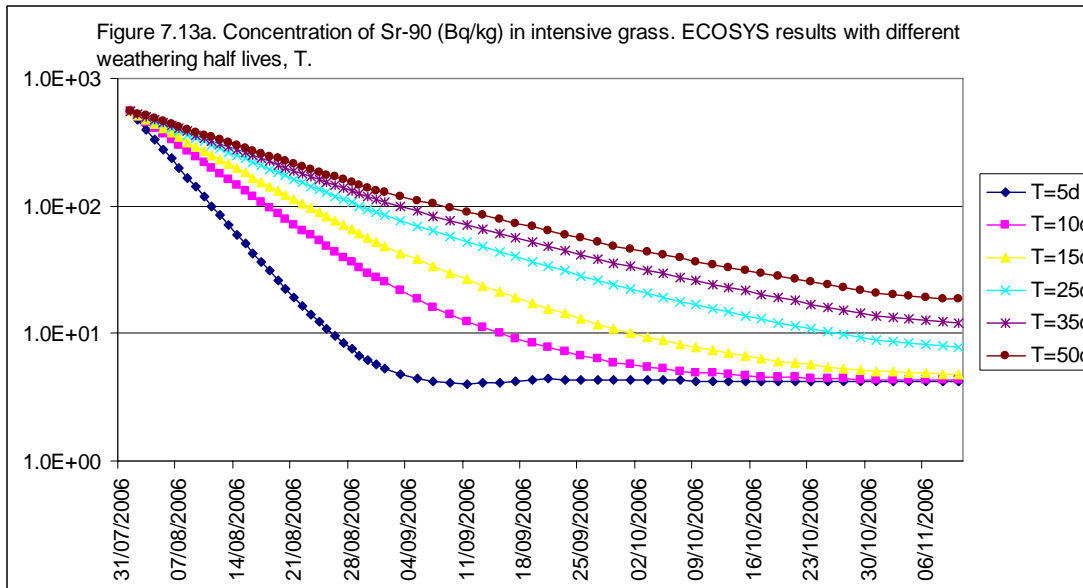
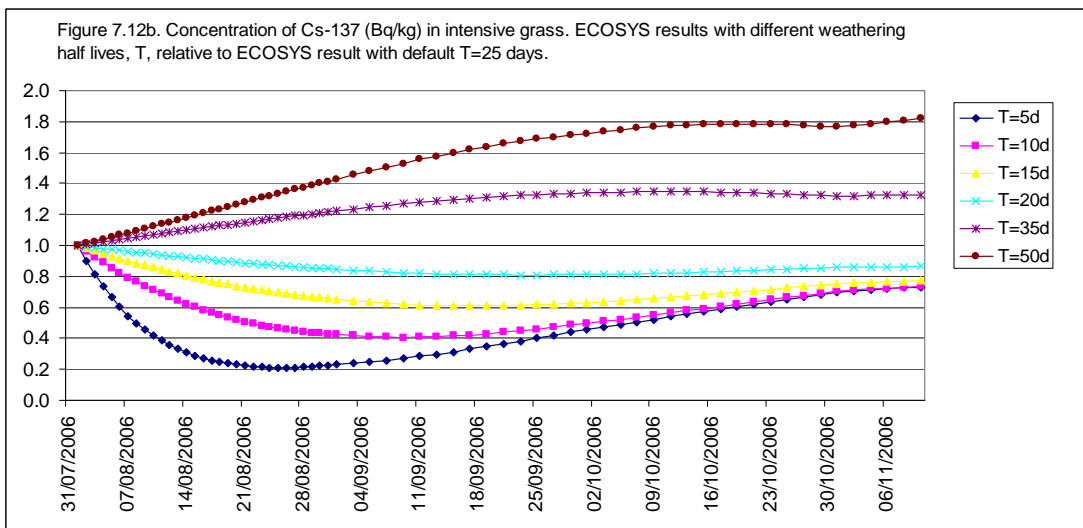
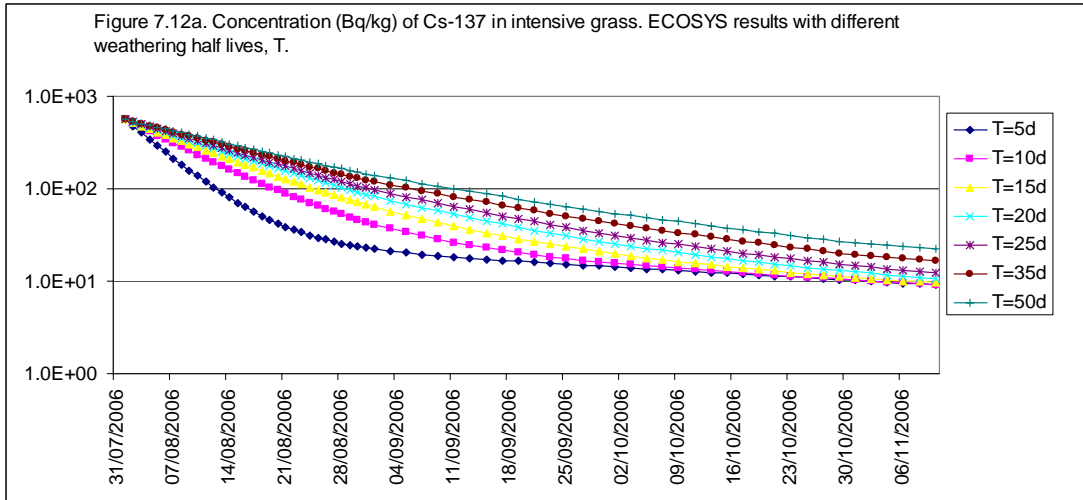
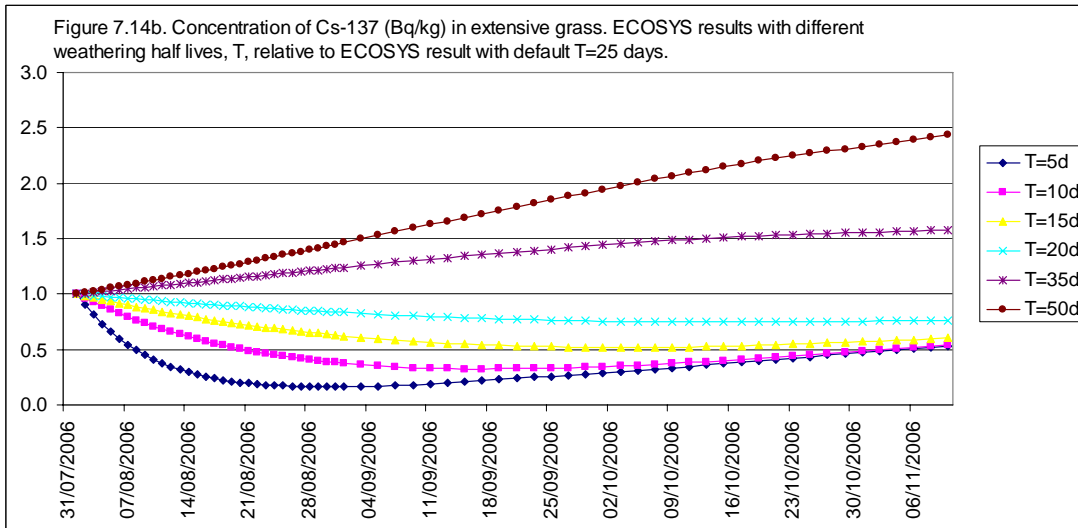
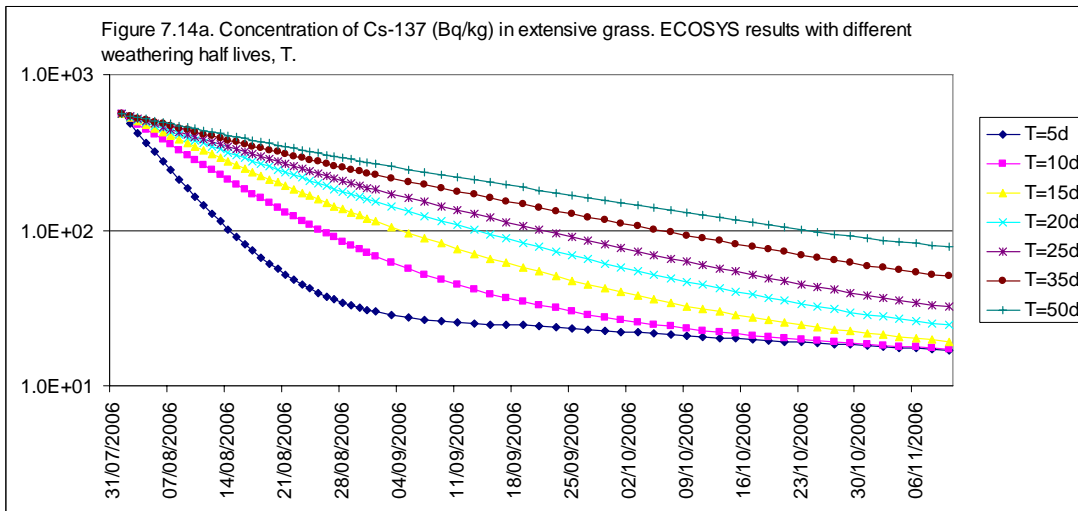
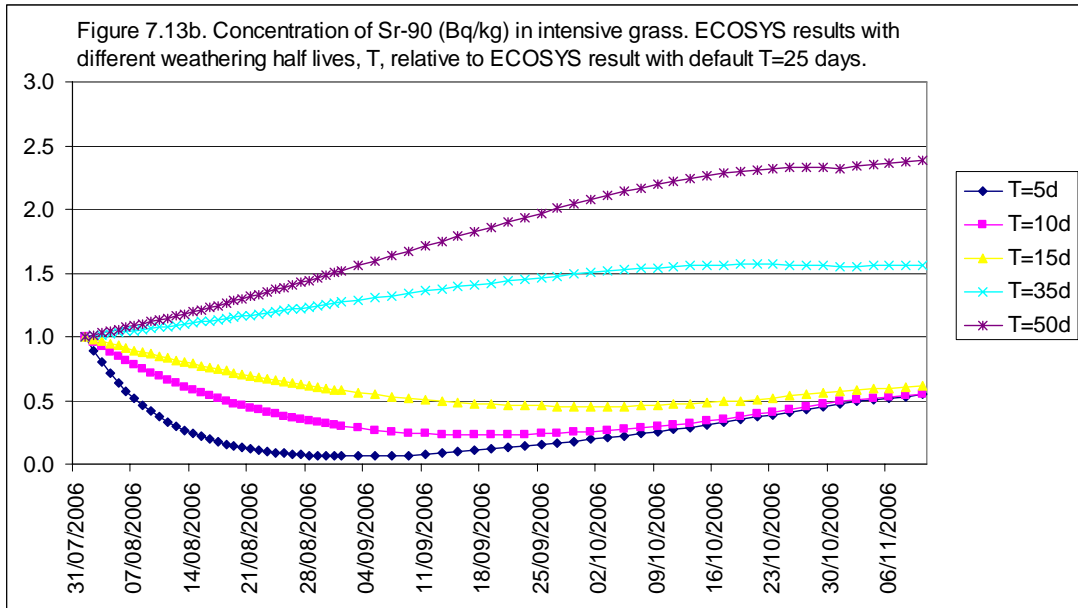


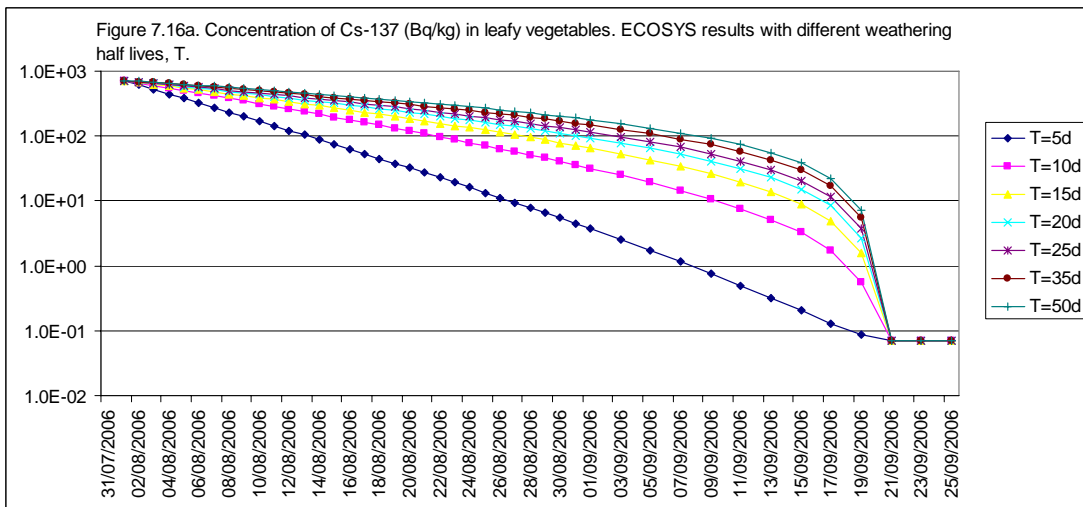
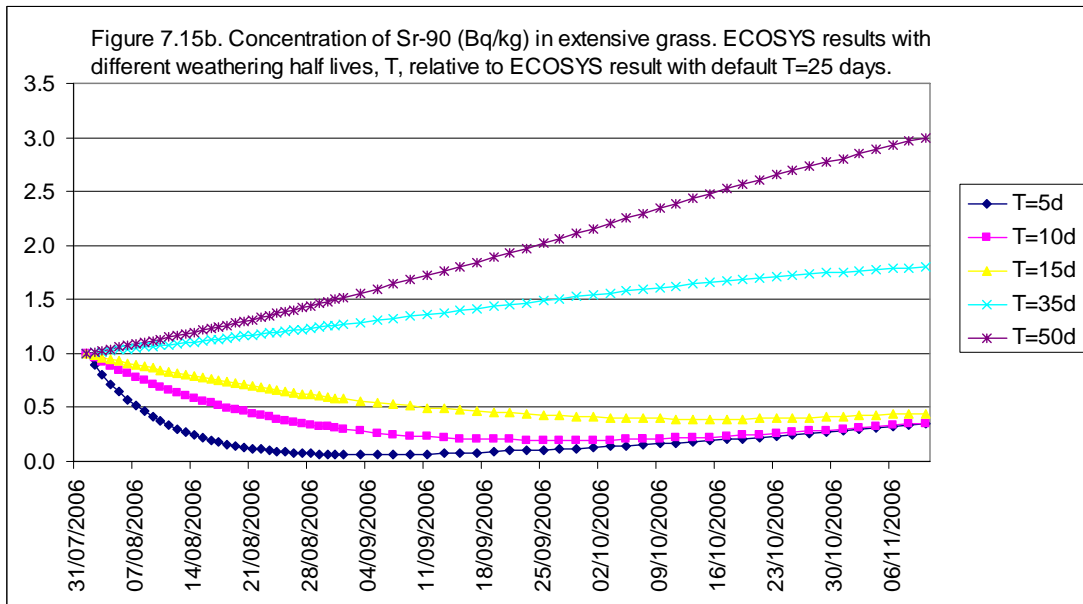
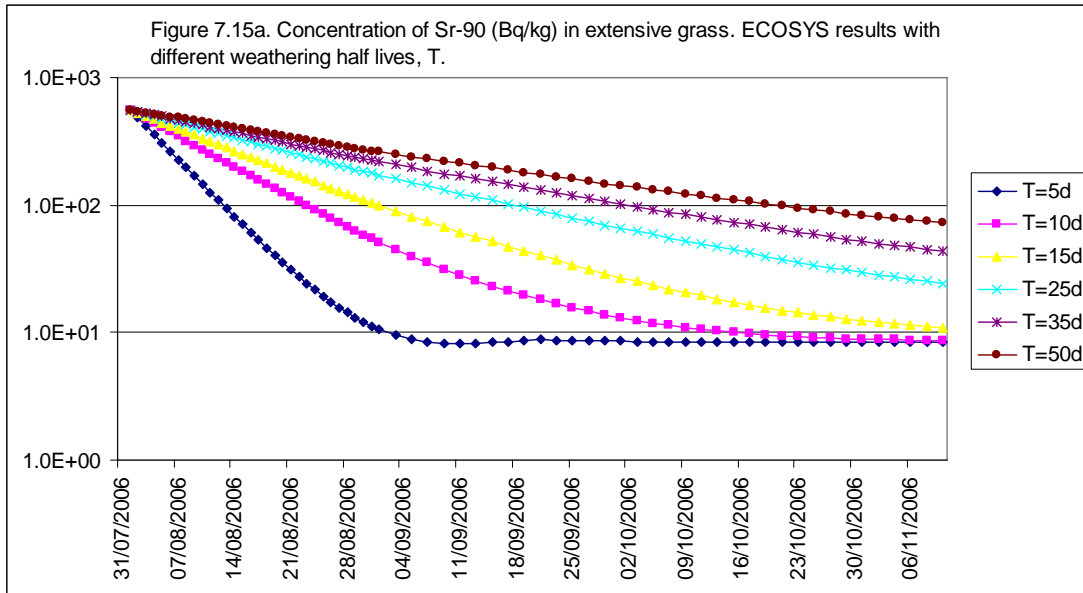
Figure 7.11b. I-131 concentration in selected products with standard and adjusted (see text) deposition velocities. The secondary y-axis shows the ratio between results with adjusted and default deposition velocities.

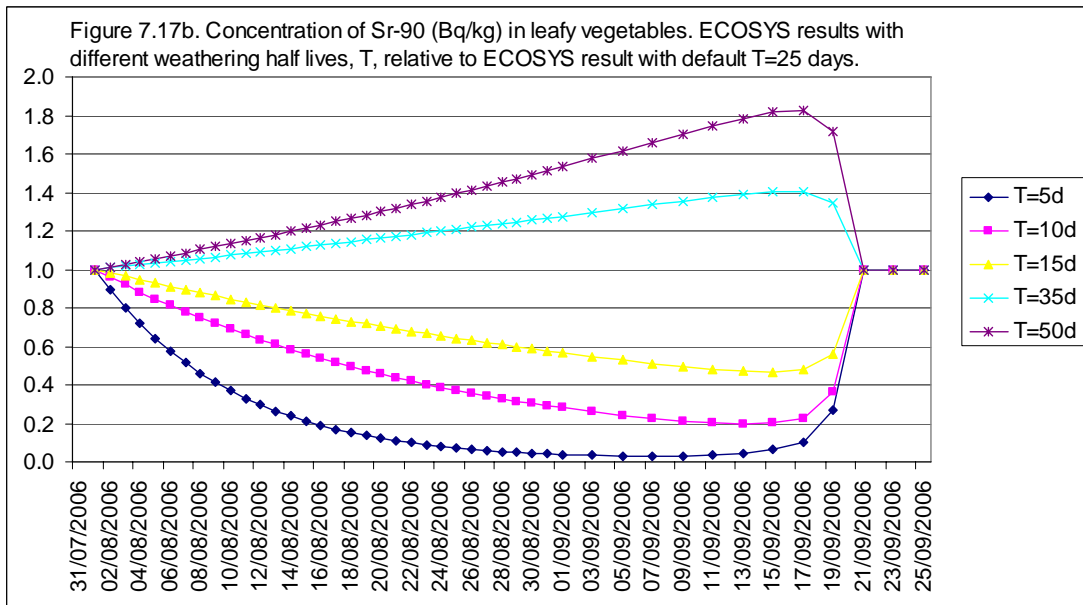
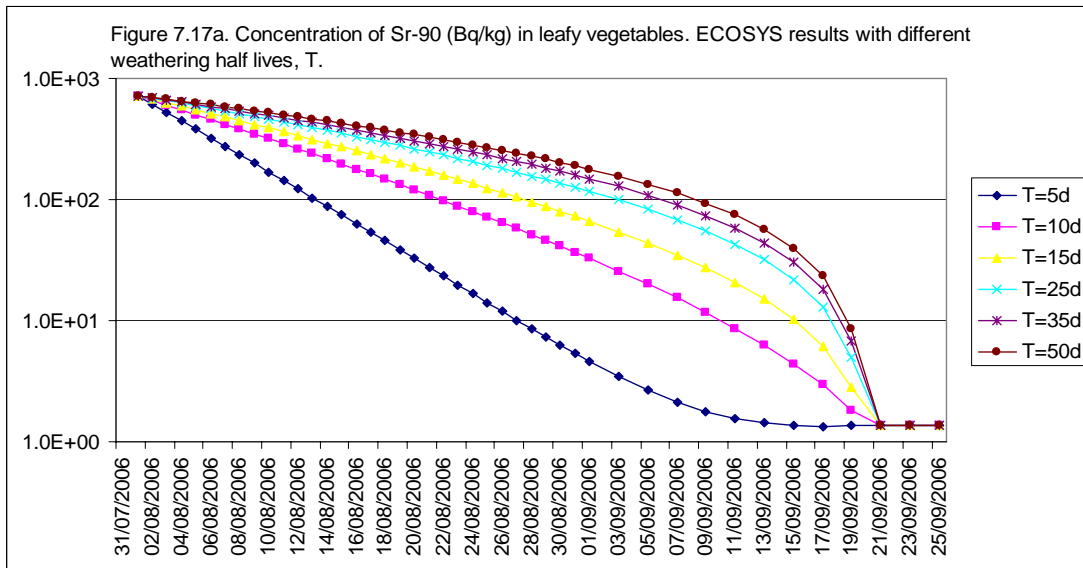
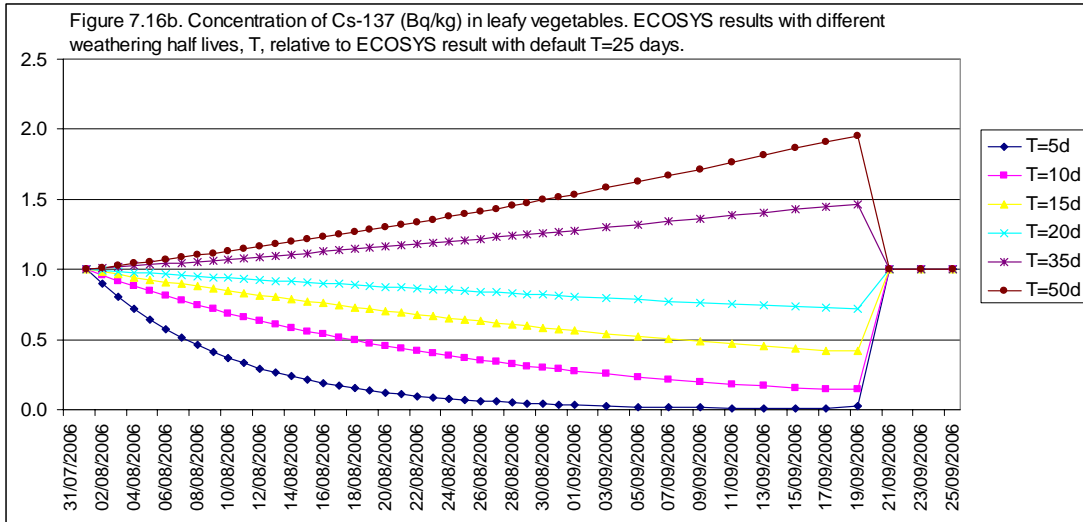


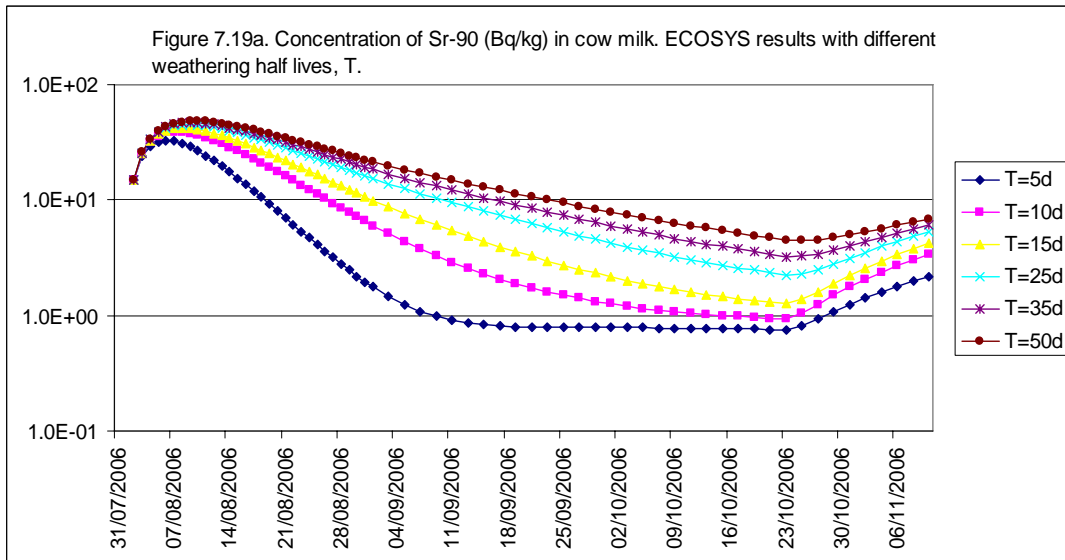
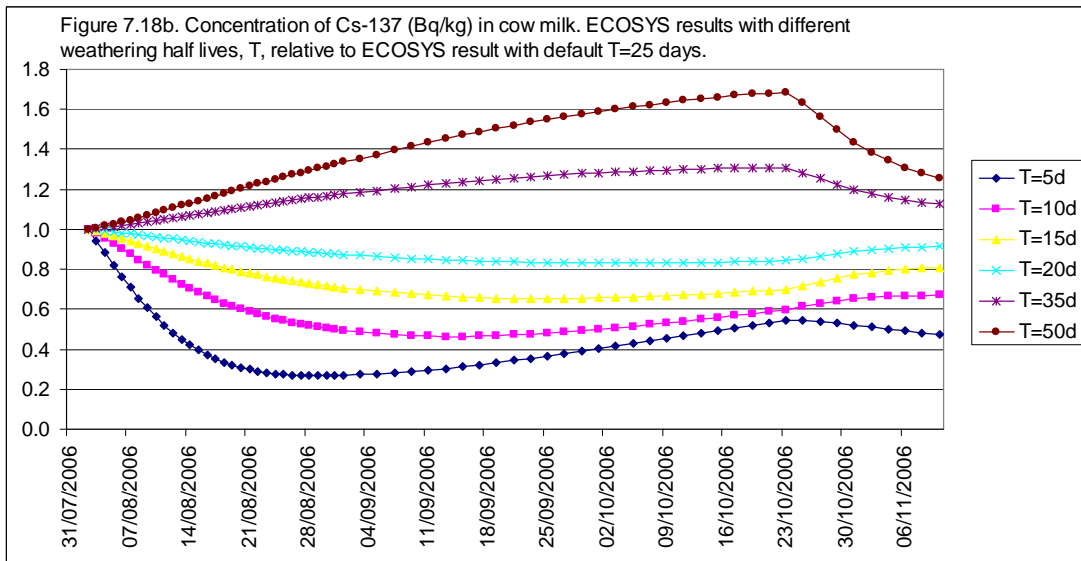
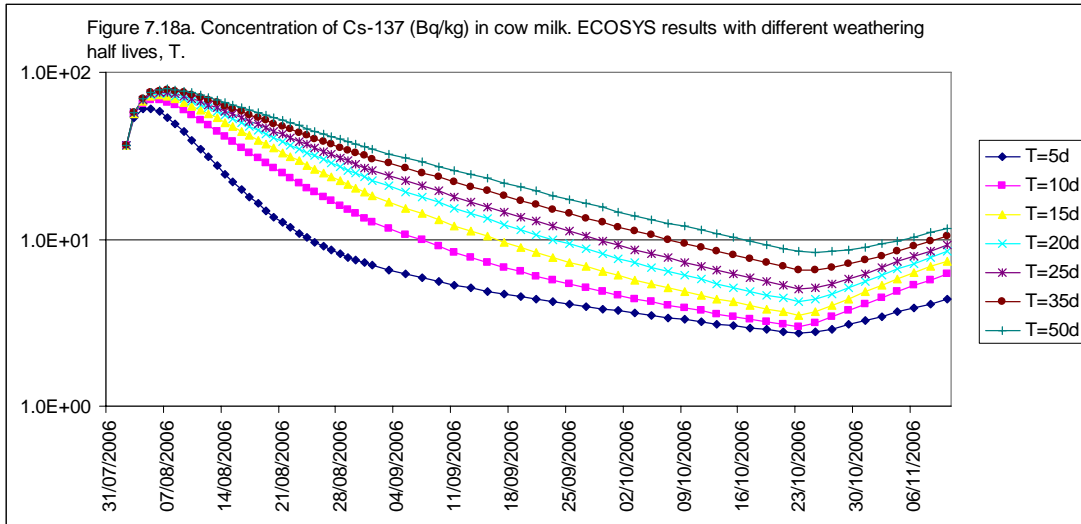


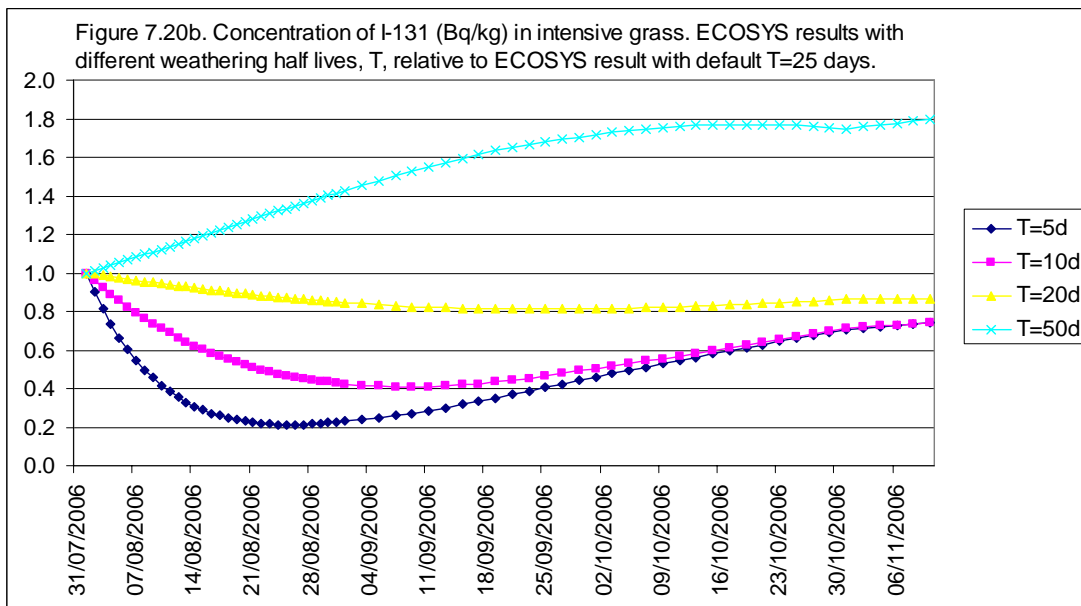
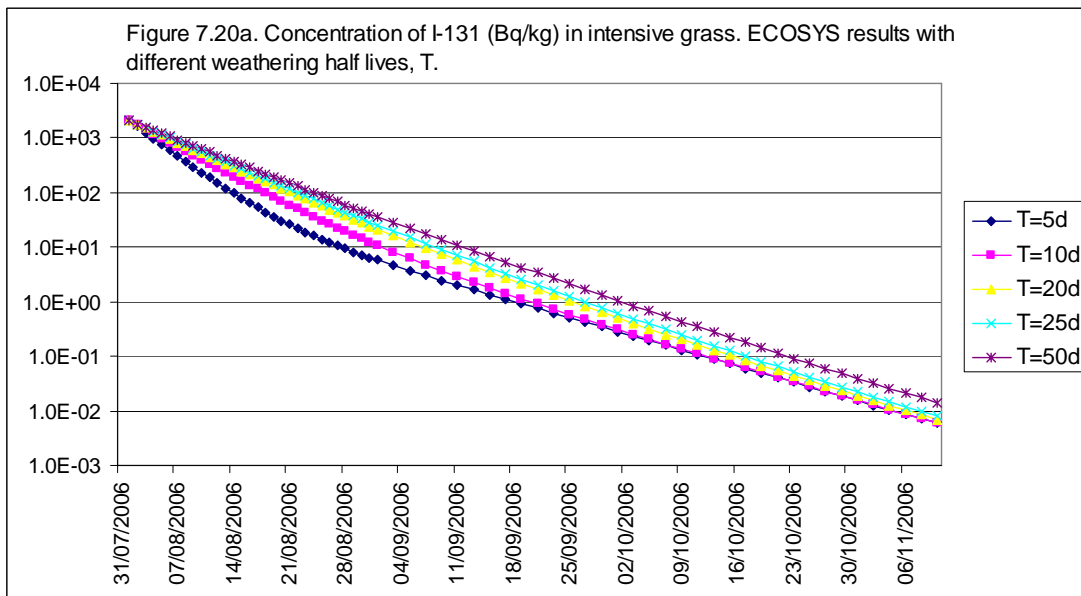
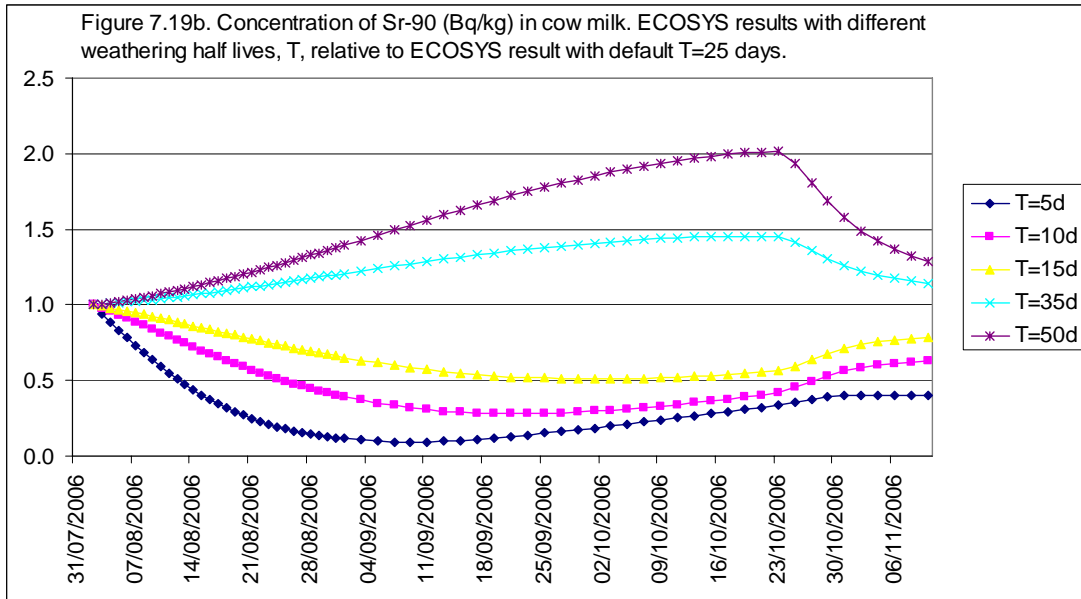


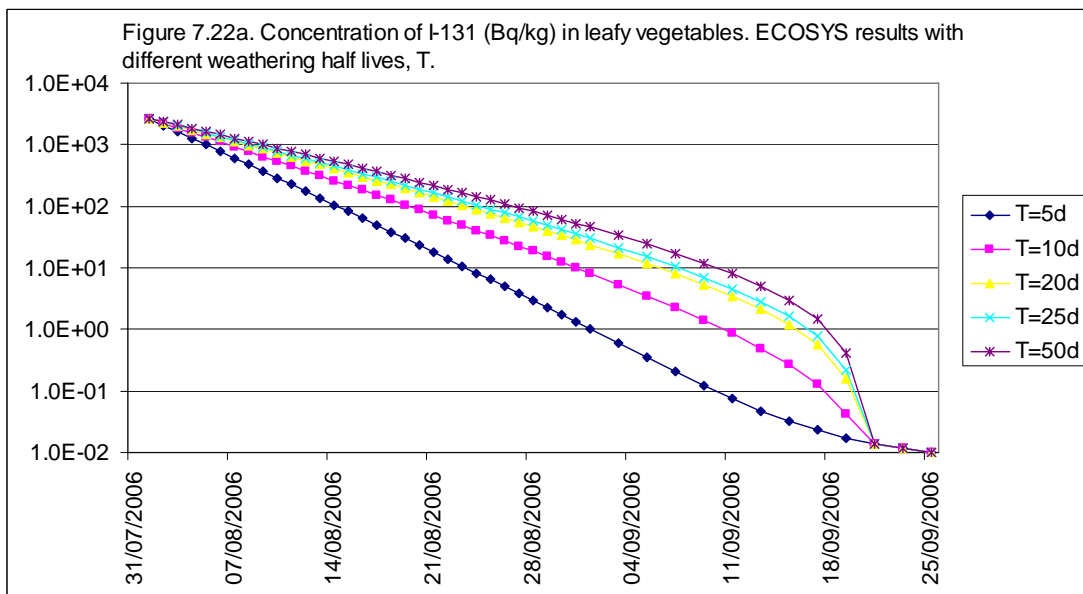
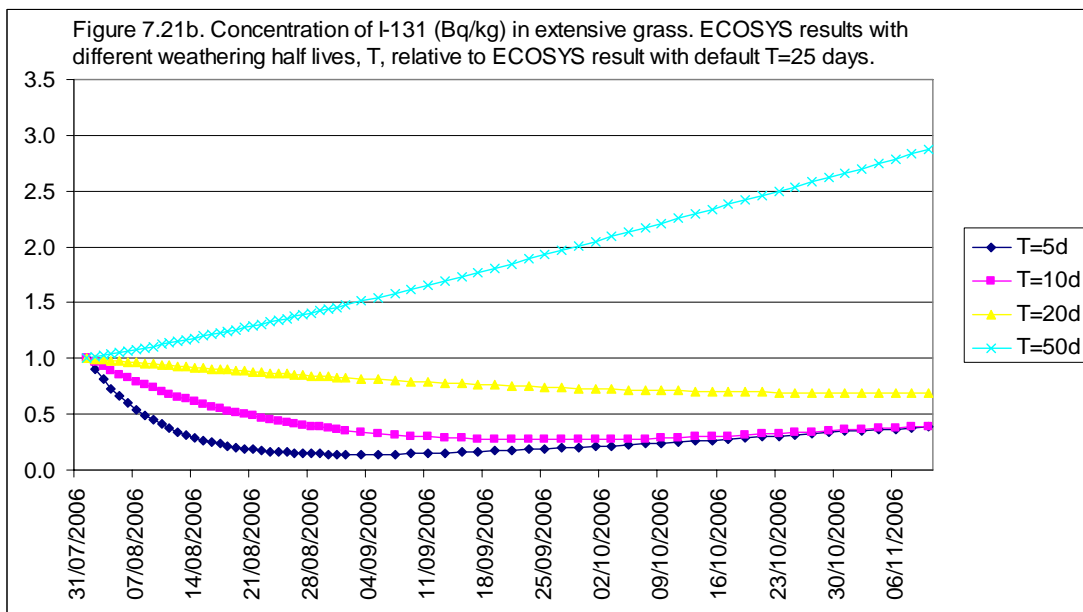
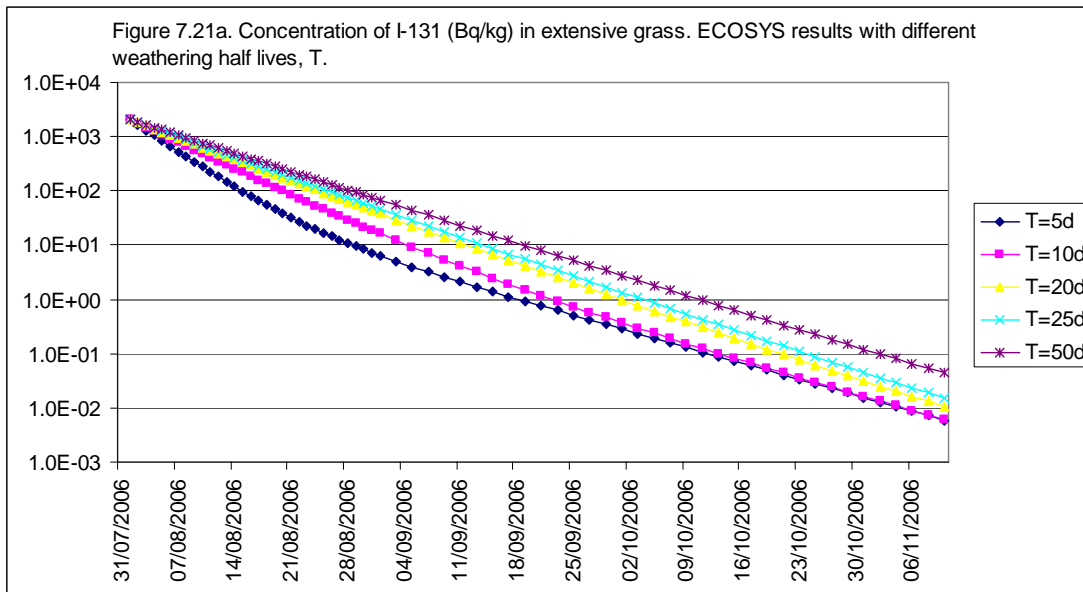


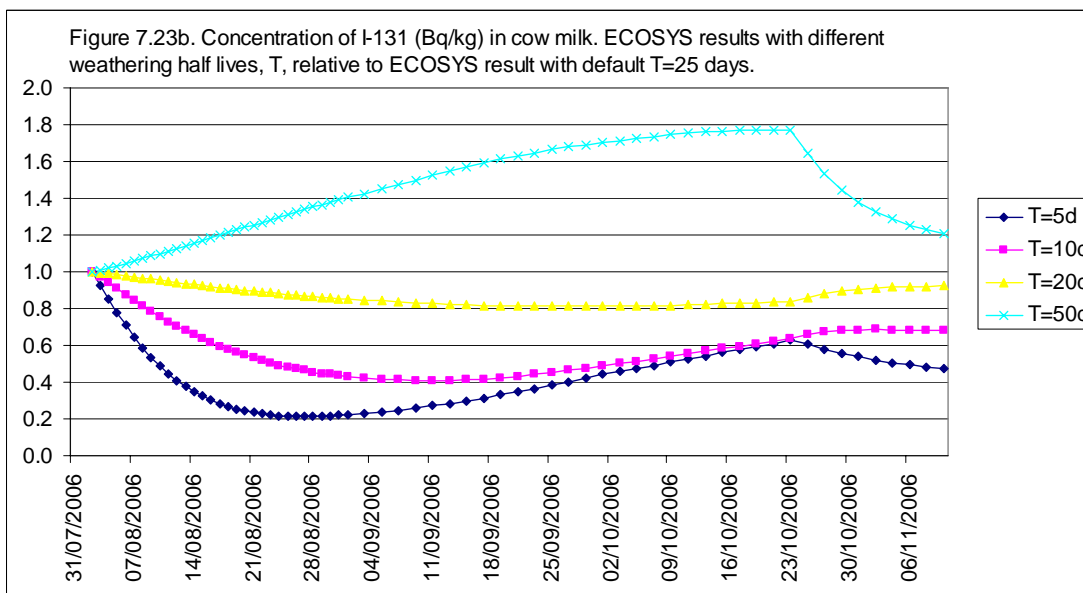
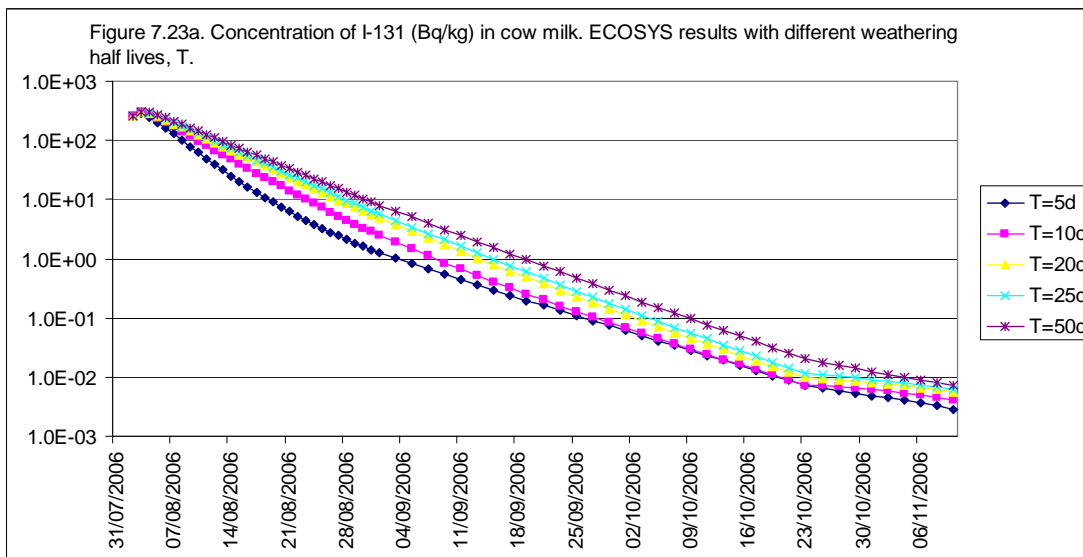
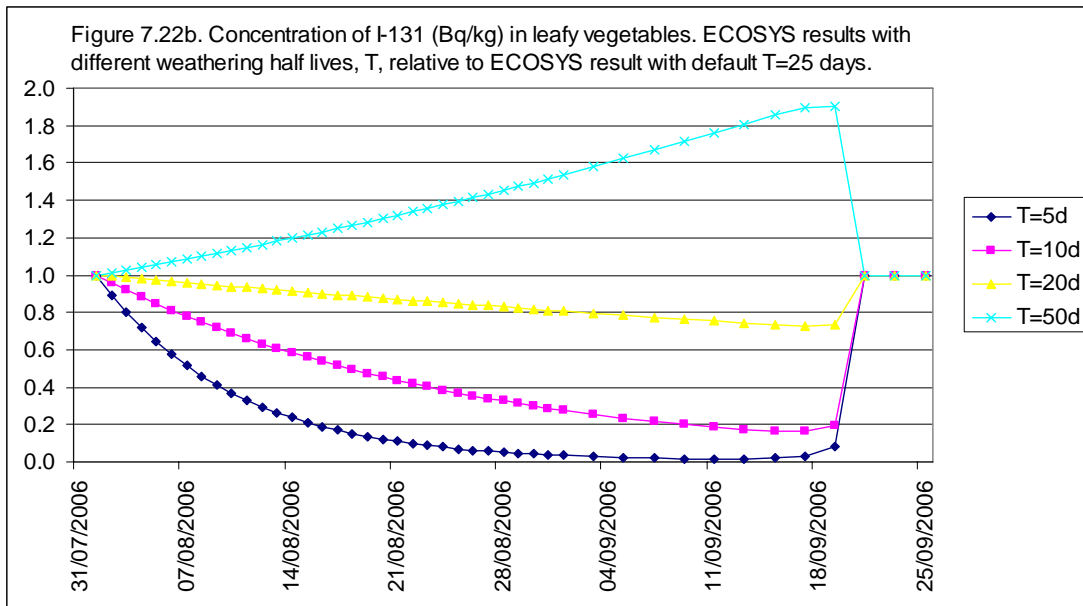












## 8. Modelling global fallout to obtain deposition time series

The global fallout from the atmospheric testing of nuclear weapons is one of the important sources of data for validating radioecological models. Often only cumulative deposition data are available, not time series that can be used for assessing results from dynamic models such as ECOSYS. Estimates of deposition time series can however be obtained from precipitation time series, which are widely available.

Previous studies have shown relationships between precipitation and deposition (Martell, 1959). This relationship has usually been expressed as the amount of deposition density (e.g. Bq m<sup>-2</sup>) in a given time period at any site in the area of study being proportional to the amount of precipitation (e.g. in mm precipitation) in the period. This is equivalent to assuming that the radionuclide concentration in the precipitation is the same during the time period in the area of study (Pálsson et al., 2006).

A linear relationship between precipitation and deposition was noted already in early radioecological studies and it was e.g. used for making an assessment of <sup>137</sup>Cs deposition in the Arctic area in the first AMAP (1998) report (Wright et al, 1999). Precipitation based estimates have been used in the Nordic countries and joint comparison of data has been carried out within the NKS-B EcoDoses and PardNor activities. The results show that the assumption holds well that the radionuclide concentration is similar in all of the Nordic region in any given time interval. This was initially tested for <sup>137</sup>Cs as reported in reports NKS-98 (Bergan & Liland, 2004), NKS-110 (Bergan et al., 2005) and NKS-123 (Nielsen and Andersson, 2006). Analysis of <sup>90</sup>Sr data shows that this holds also well for <sup>90</sup>Sr.

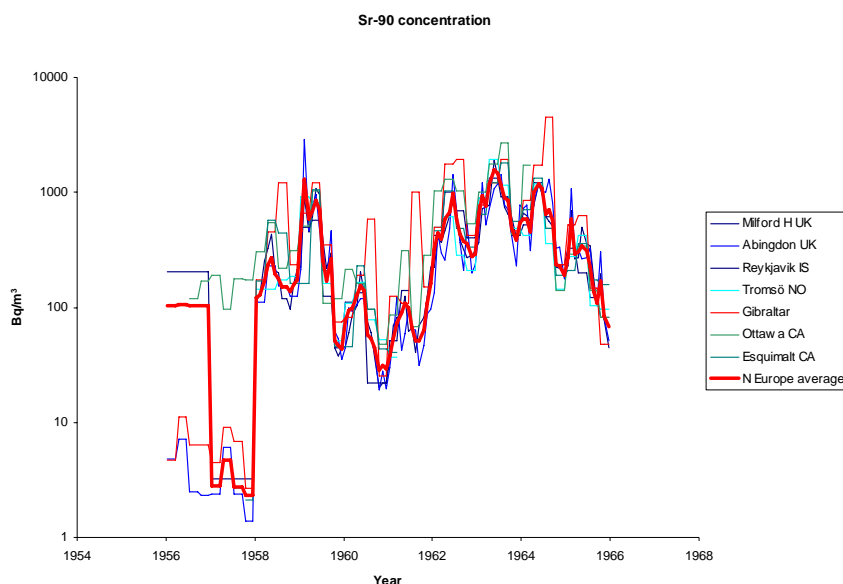


Fig. 8.1. <sup>90</sup>Sr concentration in precipitation at a few stations in Northern Europe and Northern America (Canada). The average shows a clear annual cycle.

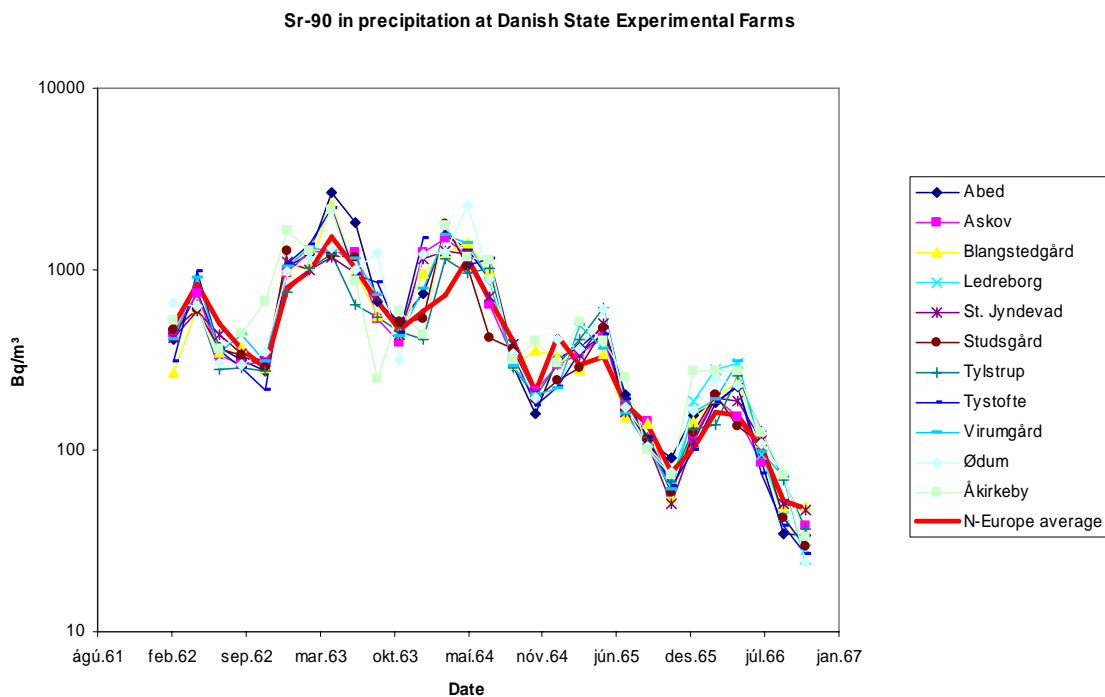


Fig. 8.2.  $^{90}\text{Sr}$  concentration in precipitation at a few stations in Denmark (data from the Risø network). Also shown is the same average as in Figure 8.1. The Risø data fit very well with the average values.

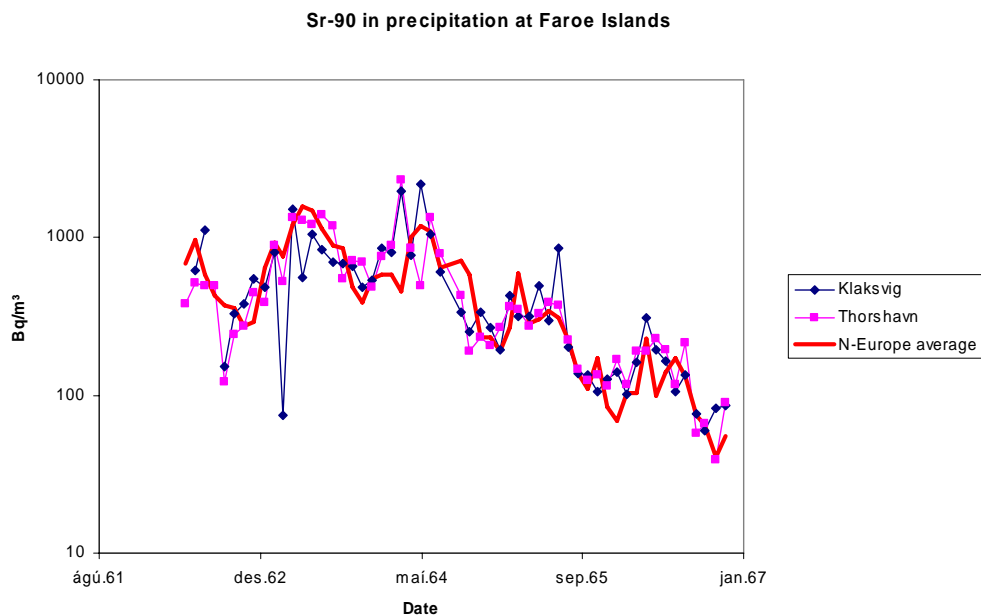


Fig. 8.3.  $^{90}\text{Sr}$  concentration in precipitation at two stations in the Faroe Islands and the Northern European average. The Faroese data fit also the average values well.



This study was followed-up in a separate NKS-B activity, DepEstimates and the final report will be published in 2011. The study used global data from the EML (US), AERE (UK) and Risø (DK) sampling networks. The main emphasis was on using the  $^{90}\text{Sr}$  data set, since it is the most complete one, but the main findings were found to be applicable for other radionuclides as well such as  $^{137}\text{Cs}$  and Pu.

It was found that latitude (and longitude) ceased to have an important explanatory power if the area of study was limited to northern latitudes, 30°- 70° N. Then much of the variability in the data could be explained by a generic temporal function (same for all sites) and the precipitation at each site.

What is interesting is that the deposition rate could be explained by a simple non-linear power function of the precipitation rate ( $p^{0.6}$ ). If the monthly precipitation was excessive, further increases had no explanatory power. It should be noted that this is a similar relationship as has been proposed (Sportisse, 2007) to link washout and rainout coefficients with rain intensity (see chapter 3 in this report).

Precipitation based deposition estimates can be of use for constructing deposition time series and improved estimates can be obtained by including precipitation rate data.

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## 9. Summary and conclusions

This year's work on the PardNor activity was targeted on the following primary investigations:

*(i) Revision of ECOSYS parameters describing deposition processes, particularly taking into account contaminant particle sizes.*

*(ii) Revision of ECOSYS parameters for description of natural weathering processes that reduce concentrations of contaminants attached to crops surfaces in the early phase after an airborne contamination has taken place.*

An overall objective was to enable integration of state-of-the-art knowledge and data in the ECOSYS model, which was parameterised about 20 years ago.

Chapter 2 of this report shows the complexity of deriving new representative dry deposition velocity parameters from experimental data, as many factors (e.g., contaminant characteristics, atmospheric conditions and surface characteristics) can strongly influence the dry deposition process. The chapter gives a systematic overview of experimental data from the literature, and suggests a new set of values for deposition to crops, bare soil and snow cover (for 'typical' atmospheric conditions), distinguishing between different particle characteristics. This data can not readily be implemented in the current version of ECOSYS (nor in ARGOS/RODOS), where the coding was made following the assumption that it is not necessary to distinguish between particle sizes in connection with the deposition modelling. However, it is clear that the ECOSYS defaults are not representative for the aerosols for which they are claimed to have been derived, and depending on the type of scenario (essentially the physicochemical forms of contaminants), estimates of contamination levels and doses based on ECOSYS default dry deposition parameters could be wrong by several orders of magnitude.

Chapter 3 suggests new methodologies for modelling the wet deposition processes washout, rainout and snow scavenging. These methodologies can not be implemented in ECOSYS, which does not include detailed modelling of wet deposition, but this could be considered for direct revision of the ARGOS and RODOS decision support systems, where the wet deposition processes are included. The chapter gives an overview of how particularly particle size, but also rain rate, affects the scavenging process.

Chapter 4 summarises a number of Nordic efforts to measure dry deposition parameters of relevance for agricultural crops and soil. Measurements were made both in Denmark, Sweden and Finland after the Chernobyl accident. Generally, the only type of crop for which Nordic measurements are available is grass. In some cases, measurements led to very high calculated deposition velocities, which could, on the basis of theory as well as other measurement sets, not exclusively be explained by dry deposition. It is however also demonstrated that the length of the grass (or in other words: the amount of vegetation per unit area) is an important factor to take into account in modelling the dry deposition processes.

Chapter 5 gives an account of the natural weathering processes that reduce levels of contaminants attached to the surfaces of crops following a contaminant deposition. Growth dilution is treated separately in ECOSYS, and therefore not included in this study. The default parameter in ECOSYS is a weathering half-life of 25 days, which is supposed to apply for all contaminants, crops and weather influences. It is clear from the data summarised in Chapter 5 that for instance weathering half-lives for elemental iodine deposition are different from those for aerosols, and that it would be advantageous to build real weather data into the modelling of the weathering process in ARGOS and RODOS, as precipitation strongly influences the weathering half-life.

Chapter 6 gives a separate account of assessments of the weathering half-life on the basis of measurements made in the Nordic countries after the Chernobyl accident. For one of the locations, the results do not seem in good agreement with results obtained elsewhere, which may indicate that growth dilution has here not been excluded in the calculations.

Chapter 7 reports on the results of a series of calculations using the ECOSYS model to demonstrate the effect of introducing the new and improved dry deposition velocity and weathering parameters that were suggested in respectively Chapter 2 and Chapter 5. It is shown that this parameter revision leads to highly significant changes in estimates of contaminant levels in food products over the first years after a 'Chernobyl scenario' contamination. The importance of revising these parameters may be even greater for other conceivable contamination scenario types.

Chapter 8 specifically discusses the dependence of the measured global fallout contamination levels on precipitation rates, suggesting a power function relationship.