



Lessons from the Fukushima and Chernobyl accidents concerning the ^{137}Cs contamination of orchard fresh fruits

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Abstract – The observations made in Japan in 2011 after the Fukushima-Daiichi accident and those made in France, Italy, Greece and Austria in 1986 after the Chernobyl fallout, show that the development stage of orchard trees at the time of atmospheric deposition is a major factor determining the level of caesium contamination in fruits at harvest. Both data sets are shown to be consistent and enable one to estimate, for mobile elements in plants such as caesium, an aggregated transfer factor (expressed in $\text{Bq}\cdot\text{kg}^{-1}$ of fresh fruit per $\text{Bq}\cdot\text{m}^{-2}$ deposited on the ground surface) whose value strongly depends on the time elapsed between fallout and harvest. The Fukushima data set also enables one to estimate effective half-lives (expressed in days), which are helpful for predicting the decrease in fruit contamination with time. We found an average value of 200 days for the one-year period after radioactive fallout, which is quite consistent with values estimated from post-Chernobyl surveys.

Keywords: Fukushima-Daiichi NPP accident / Chernobyl / fruit contamination / caesium dynamics / transfer parameters

1 Introduction

Since the occurrence of the Fukushima nuclear accident, hundreds of thousands of pieces of data on foodstuff contamination have been published by the Japanese Ministry of Health, Labor and Welfare on their website (MHLW, 2013). Those concerning caesium radioisotopes in vineyard grapes and orchard fruits such as apricots, cherries, peaches, apples and pears, are of particular interest. Indeed, the existing models dealing with radionuclide transfer to fruits following an accidental atmospheric deposition are still based on a very limited number of data (see, for example, Müller and Pröhl, 1993; Brown and Simmonds, 1995; IAEA, 2003; Brown and Sherwood, 2012). After the Chernobyl accident, the few field surveys were mainly carried out in southern European countries, *i.e.* Greece, Italy and France (Silva *et al.*, 1989; Monte *et al.*, 1990; Antonopoulos-Domis *et al.*, 1991; Anguissola Scotti and Silva, 1992; Antonopoulos-Domis *et al.*, 1996; Carini *et al.*, 1996; Carini and Lombi, 1997; Madoz-Escande *et al.*, 1997, 1998; Carini, 1999; Carini and Bengtsson, 2001; Renaud *et al.*, 2003b). These did not provide enough quantitative information to enable the development of dynamic modeling approaches or estimate robust parameter values, *e.g.* kinetic rates or time-dependent transfer functions.

It is worth noting that all of these predictive models would have strongly underestimated caesium concentrations in Japanese fruits, because all of them neglect the foliar

contamination pathway at the earliest development stages of the plant cycle. The situation was such for most of the fruit species that were collected in the Fukushima region, as neither the leaves nor the flowers were formed at the time of the fallouts (mid-March 2011) (Takata, 2013). Contamination of Japanese fruit was significant, reaching some tens to some hundreds of $\text{Bq}\cdot\text{kg}^{-1}$ of fresh weight, and was still measurable in 2012 for most fruits. However, compared with the significance of the radioactive fallouts (MEXT, 2013), fresh fruits were relatively preserved due to the earliness of the accident. To some extent, this was also the case in France in 1986, despite the fact that the Chernobyl accident occurred later in the season (beginning of May 1986). The data presented below allow deducing empirical transfer factors for the first year in the particular context of this accident, as well as effective half-lives of decrease in ^{137}Cs activity levels during the following two years. This knowledge is then completed and compared with that deduced from observations made after the Chernobyl accident.

2 Contamination of fruits at first harvest

2.1 Fukushima data

For the six fruit species considered, Table 1 provides mass activities of fresh fruits at harvest in $\text{Bq}\cdot\text{kg}^{-1}$ fresh weight, for selected municipalities of the Fukushima Prefecture. The selection includes areas where both ^{134}Cs and ^{137}Cs

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Table 1. $^{134+137}\text{Cs}$ initial deposits and activities in fruits from Fukushima Prefecture.

Fruits	Municipality ^(a)	Mean deposit kBq.m ⁻²	Number of samples	Mean activity (min-max) Bq.kg ⁻¹ fw	T_{ag} m ² .kg ⁻¹ fw	2011		2012		2013	
						Number of samples	Mean activity (min-max) Bq.kg ⁻¹ fw	Number of samples	Mean activity (min-max) Bq.kg ⁻¹ fw	Number of samples	Mean activity (min-max) Bq.kg ⁻¹ fw
Apricots	Kori-machi	101	12	320 (33–330)	3×10^{-3}	3	40 (33–46)	22	30 (5.2–89)		
	Fukushima-shi	101	15	210 (63–690)	2×10^{-3}	0	–	19	26 (10–63)		
	Date-shi	99	19	350 (110–760)	3×10^{-3}	0	–	27	26 (6.4–65)		
	Minamisoma-shi	83	4	530 (250–750)	6×10^{-3}	0	–	0	–		
	Koriyama-shi	81	2	239 (210–270)	3×10^{-3}	6	39 (10–46)	2	6.5 (4.9–8.1)		
	Kunimi-shi	64	4	200 (36–340)	3×10^{-3}	4	82 (40–210)	5	15 (11–25)		
	Tamura-shi	51	4	150 (88–400)	3×10^{-3}	3	23 (8–45)	0	–		
	Soma-shi	47	2	290 (160–420)	6×10^{-3}	15	34 (5–29)	6	13 (5.7–20)		
	Iwaki-shi	30	2	95 (77–110)	3×10^{-3}	5	25 (9–38)	3	9.8 (6.1–12)		
	Takasaki-shi	15	5	23 (20–28)	2×10^{-3}	1	5	0	–		
Mito-shi	5.7	2	37 (33–40)	6×10^{-3}	0	–	0	–			
Cherries	Date-shi	99	1	66	7×10^{-4}	7	22 (11–41)	3	13 (7–17)		
	Fukushima-shi	101	10	84 (70–96)	8×10^{-4}	2	15 (14–15)	1	11		
	Kori-machi	101	1	91	9×10^{-4}	5	25 (16–35)	1	26		
	Kunimi-machi	64	1	68	1×10^{-3}	6	20 (8–38)	2	10		
Peaches	Date-shi	99	45	47 (16–160)	5×10^{-4}	11	13 (8–30)	–	–		
	Fukushima-shi	101	60	45 (16–72)	4×10^{-4}	17	8 (4–13)	–	–		
	Kori-machi	101	23	50 (23–94)	5×10^{-4}	5	15 (7–31)	–	–		
	Kunimi-machi	64	20	33 (18–58)	5×10^{-4}	2	14 (10–18)	–	–		
Apples	Date-shi	99	14	42 (24–90)	4×10^{-4}	5	14 (8–18)	–	–		
	Fukushima-shi	101	38	32 (17–39)	3×10^{-4}	21	9 (7–13)	–	–		
	Kori-machi	101	4	73 (39–62)	7×10^{-4}	4	13 (8–18)	–	–		
Pears	Fukushima-shi	101	17	23 (14–36)	2×10^{-4}	12	8 (6–12)	–	–		
	Minamisoma-shi	83	15	29 (12–46)	3×10^{-4}	12	10 (5–13)	–	–		
Grapes	Kori-machi	101		24 (23–24)	2×10^{-4}	2	6 (6–6)	–	–		
	Date-shi	99		27 (31–35)	3×10^{-4}	9	9 (5–13)	–	–		
	Fukushima-shi	101		26 (18–41)	3×10^{-4}	14	7 (3–11)	–	–		

(a) All municipalities are situated in the eastern part of the Fukushima Prefecture apart from Takasaki-shi and Mito-shi, which belong to Gunma and Ibaraki Prefectures, respectively.

concentrations were above the detection limits in 2011. The values correspond to the measured contamination at the date of sampling (harvest), thus including the radioactive decay from the deposition date. When the ^{134}Cs activity was under the detection limit (sometimes for the 2012 or 2013 harvests), it was calculated from the ^{137}Cs activity considering an isotopic ratio equal to that at the date of the deposits. The different fruit species were gathered at varying dates ranging from May–June 2011 for apricots and cherries to September–November for apples and pears. Peaches and grapes were ready for picking in July–August and early fall, respectively. The activity of $^{134+137}\text{Cs}$ deposited onto the ground surface (Bq.m^{-2}) was estimated from airborne gamma-ray monitoring and dose rate checking 1 m above the ground ($\mu\text{Sv.h}^{-1}$). The surveys were regularly operated by the Ministry of Education, Culture, Sports, Science and Technology (MEXT, 2013) from April 2011, in the Fukushima Prefecture and neighboring Prefectures. The values were converted by the authors as of March 15, 2011, by applying a physical decay correction factor. As displayed in Table 1, a mean deposit

was evaluated for each municipality, by spatially averaging the ground surface deposit measured during the so-called “fourth airborne survey” in November 2011. Over 80% of the deposits occurred with rain on Date, Fukushima, Koriyama or Kori. In Minamisoma or Kawamata the wet fraction ranged from 40% to 90%, whereas in Soma the deposits were mainly dry. For all species, the highest activities were generally observed in the highly contaminated areas, such as Kori-machi, Fukushima-shi and Date-shi, where the mean deposit was about 100 kBq.m^{-2} . On the contrary, the contamination level in (apricot) fruits originating from less affected areas was significantly lower, such as in Iwaki, Takasaki-shi and Mito-shi counties. Thus, some correlation exists between caesium concentration in fruits, at first harvest, and ground surface deposit, despite the observed variability. This variability is much less pronounced when normalizing the activity in fruit by the local mean deposit, which is expressed in $\text{Bq.kg}^{-1} \text{ fw}$ per Bq.m^{-2} . This normalized activity is usually known as the aggregated transfer factor, T_{ag} ($\text{m}^2.\text{kg}^{-1} \text{ fw}$). From Table 1, it can be observed that the spatial variability of TF, especially for apricot, has been considerably

Table 2. ^{137}Cs initial deposits and activities in fruits measured in 1986 in Italy and France following the Chernobyl accident.

Fruits	Location	Deposit kBq.m ⁻²	Number of samples	Mean activity min-max) Bq.kg ⁻¹ fw	T_{ag} m ² .kg ⁻¹ fw	Reference
1986						
Apples	Northern Italy 2	15.1 ^(a)	6	43 (30–64)	3×10^{-3}	Anguissola Scotti and Silva, 1992
	Central Western Italy	n.a.	6	n.a.	3×10^{-3}	Monte <i>et al.</i> , 1990
	Southeastern France	n.a.	n.a.	n.a.	4×10^{-3}	Maubert and Roussel, 1988
	Southeastern France	n.a.	n.a.	n.a.	2×10^{-3}	Maubert and Roussel, 1988
Apricots	Southeastern France	n.a.	n.a.	n.a.	1×10^{-2}	Maubert and Roussel, 1988
Pears	Northern Italy 2	15.1 ^(a)	5	43 (35–48)	3×10^{-3}	Anguissola Scotti and Silva, 1992
	Southeastern France	n.a.	n.a.	n.a.	3×10^{-3}	Maubert and Roussel, 1988
Peaches	Northern Italy 1	7.5 ^(a)	5	22 (19– 28)	3×10^{-3}	Anguissola Scotti and Silva, 1992
	Northern Italy 2	15.1 ^(a)	2	39 (21– 47)	3×10^{-3}	Anguissola Scotti and Silva, 1992
	Southeastern France	n.a.	n.a.	n.a.	1×10^{-2}	Maubert and Roussel, 1988
Cherries	Northern Italy 3	7.2 ^(a)	3	30 (23– 36)	5×10^{-3}	Anguissola Scotti and Silva, 1992
	Southeastern France	n.a.	n.a.	n.a.	3×10^{-2}	Maubert and Roussel, 1988
Grapes	Northern Italy	14.8	21	2.4 (1.3– 5)	2×10^{-4}	Silva <i>et al.</i> , 1989
	Central Italy	3.7	9	1.4 (0.8– 2.2)	4×10^{-4}	Silva <i>et al.</i> , 1989
	Southern Italy	0.7	4	2.4	3×10^{-3}	Silva <i>et al.</i> , 1989
Sour cherries	Northeastern France 1	4.4	2	120 (50– 180)	3×10^{-2}	SCPRI, 1986
	Northeastern France 2	10.5	1	300	3×10^{-2}	SCPRI, 1986
	Northeastern France 3	6.0	5	140 (60–170)	2×10^{-2}	SCPRI, 1986

(a): evaluated from soil massic activity measurements assuming a soil density of 1500 kg.m⁻³.

lowered. One can note that TF values are apparently (two times) higher than average in low-elevation coastal areas, *i.e.* in Minamisoma-shi and Soma-shi municipalities, located to the North of the nuclear site, and Mito-shi to the South in Ibaraki Prefecture. This could result from the more advanced growth stage of apricot trees, probably in the flowering stage in March 2011. Despite this normalization, significant differences apparently remain among some fruit species. Apricots are characterized by the highest transfer factor values, ranging between 2×10^{-3} and 6×10^{-3} m².kg⁻¹ fw. T_{ag} values are obviously lower for fruit species such as apples or pears that are harvested in late fall or early winter, and vary from 2×10^{-4} to 7×10^{-4} m².kg⁻¹ fw. The foliar transfer factor differs by more than one order of magnitude between these two extremes. These observations seem to corroborate the dependence of the foliar transfer upon the vegetation development stage at the time of deposition. A part of the variability observed in the T_{ag} values among municipalities and/or species could also originate in the characteristics of atmospheric deposition, as the fraction of deposited caesium aerosols that is intercepted by vegetation is known to strongly depend on wind and rainfall characteristics. This point will be discussed later.

2.2 Chernobyl data

After the Chernobyl accident, measurements of contamination in fruits harvested in 1986 were very scarce, and even fewer were those which enabled one to estimate foliar transfer factor values. Table 2 provides a few measurements of ^{137}Cs activities in tree fruits, sampled either in Italy or

France (SCPRI, 1986; Maubert and Roussel, 1988; Silva *et al.*, 1989; Monte *et al.*, 1990; Anguissola Scotti and Silva, 1992). Grape samples were collected in northern, central and southern Italy together with the deposited activities reported by the authors for each geographical area. In these southern European countries, these different fruit species are usually gathered at characteristic dates varying from June–July for apricots and cherries to fall for apples and pears. Peaches and grapes are usually collected in July–August and August–September, respectively. The deposits were mainly wet (over 80%). If we exclude the outsider value from Maubert and Roussel (1988), the T_{ag} for apples, pears and peaches feature quite similar values, ranging from 2×10^{-3} to 4×10^{-3} m².kg⁻¹ fw. T_{ag} for cherries are slightly higher (5×10^{-3} m².kg⁻¹ fw), this being linked perhaps to their earlier development stage at the time of deposition. The transfer factors obtained by Maubert and Roussel, (1988) for cherries, apricots and peaches range from 1×10^{-2} to 4×10^{-2} m².kg⁻¹ fw. Considering that atmospheric deposition occurred with a rainfall amount of from 5 to 15 mm and setting the harvest date to July 1 (*i.e.* 2 months after deposition), the foliar transfer factor values calculated by the ECOSYS model are quite similar, ranging from about 2×10^{-2} to 6×10^{-2} m².kg⁻¹ fw.

2.3 Discussion

Figure 1 synthesizes all the TF values estimated at first harvest from Fukushima and Chernobyl field measurements described above. Values are displayed against the time elapsed between deposition and harvest dates. The deposition dates are

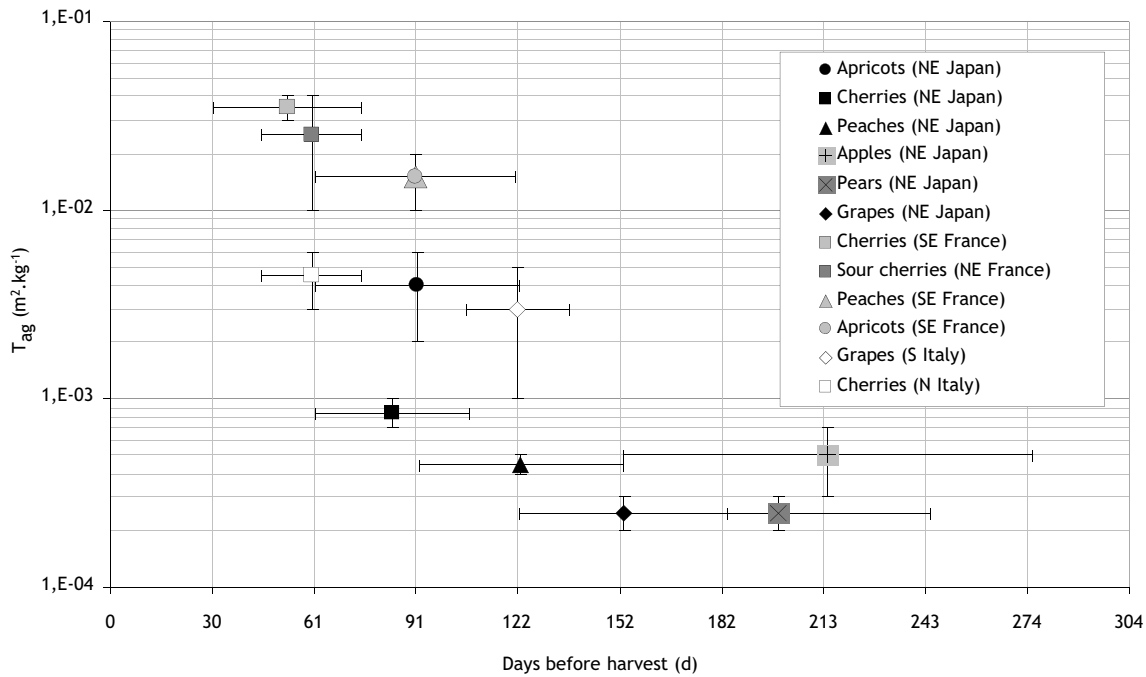


Fig. 1. Foliar aggregated transfer factor at first harvest, T_{ag} ($\text{m}^2 \cdot \text{kg}^{-1} \text{fw}$), as a function of time elapsed since deposition, for orchard tree fruits contaminated by Chernobyl and Fukushima field deposits. Error bars cover the min-max range of variation for both harvesting period and T_{ag} values. Dot points indicate the average value between min and max.

fixed to May 1 and March 15 for Chernobyl and Fukushima, respectively. Uncertainty in T_{ag} values and harvest dates are accounted for here.

However, we must recognize that some variability remains among fruit species and geographical areas, *i.e.* up to one order of magnitude for harvest time between 2 and 4 months after the fallout. This variability is induced by relatively well-known environmental factors, at least for annual crops, that are now discussed.

1. First of all, we must keep in mind that the foliar transfer factor, defined here as the fruit contamination at harvest date t normalized by ground deposit at date of accident t_a , implicitly includes the variability of the interception factor, IF (m^2 of foliar surface per m^2 of soil). This factor is defined as the fraction of the deposited contamination that is effectively captured by the orchard tree canopy at time t_a . Actually, $T_{ag}(t)$ can be expressed as follows:

$$T_{ag}(t) = IF(t_a) \times T_{fr}(t - t_a) \times \exp(-\lambda(t - t_a))$$

where $T_{fr}(t - t_a)$ is the true translocation factor (in $\text{m}^2 \cdot \text{kg}^{-1} \text{fw}$) and λ (day^{-1}) represents the physical decay. This last term accounts for the radionuclide decay from deposition to harvest. The interception of airborne caesium aerosols by aerial vegetation is known to be much less effective when deposition occurs during a rainy episode, and interception factors are usually greater for dry deposition conditions. This is why most predictive models propose higher T_{ag} values for dry deposition, as in the ECOSYS (Müller and Pröhl, 1993), FARM-LAND (Brown and Simmonds, 1995) and ASTRAL models (Renaud *et al.*, 1997). Moreover, IF values are known to decrease with an increase in rainfall height (see, for example, Pröhl, 2009). In the ASTRAL model, the tabulated T_{ag} values

for various agricultural crops decrease by 3 to 10 times when the rainfall amount increases from 5 to 15 mm. Antonopoulos-Domis *et al.* (1991) studied the contamination of fruit on two Greek sites from 1987 to 1990: the Thessaloniki and Naoussa sites, where ^{137}Cs deposits induced by rain were, respectively, equal to 15 and 63 $\text{kBq} \cdot \text{m}^{-2}$. Noting that the contamination of cherries, apricots and pears was at the same level on both sites and even higher for cherries and apricots from the Thessaloniki site, they investigated two new sites mainly affected by dry deposition: Xanthi and Ionnina, where deposits were about 2.5 $\text{kBq} \cdot \text{m}^{-2}$. Despite such a difference in ground surface inventory between wet and dry contaminated sites, they observed that activity in leaves of cherry trees from Naoussa (wet) was only 3 to 5 times higher than those from Xanthi (dry) and Ionnina, Thessaloniki being an intermediate case.

Radioactive fallouts in western European countries after the Chernobyl accident were mainly due to precipitation, and were shown to increase with the rainfall amount (Renaud *et al.*, 2003a). It was notably the case for the various areas where fruit samples came from. The proportion of wet deposited caesium was estimated to vary between 80% and more than 90%. The situation was much less clear in Japan. It was shown that in some regions, the contribution of dry deposition could be significant, especially to the South-Southeast and to the North of the nuclear site (Katata *et al.*, 2012a, 2012b; Terada *et al.*, 2012; Champion *et al.*, 2013; Korsakissok *et al.*, 2013). This variability in the atmospheric deposition conditions may play a significant role in the dispersion of TF values, as observed in Figure 1.

2. Second, we must keep in mind that the deposit-to-harvest time delay remains a poor approximation of the vegetation stage at the time of deposition. Notably, it cannot represent the variability that may be observed in a crop development

stage due to the variability of soil and climate conditions, or even agricultural practices. An example is given by the data of Silva *et al.* (1989) in Table 2. TF values for grapes increase by one order of magnitude from North to South Italy. The authors explain that in northern Italy, the vines had already budded in April 1986 but not yet flowered, whereas in southern Italy, vines were ahead by 15 to 20 days and already flowering or very close to this stage. It must be noted that the vine produces its leaves before flowering, unlike apricot and cherry trees. This spatial variability of the development stage might explain the difference observed in apricot samples collected from either coastal or mountainous production areas in Japan (see Tab. 1). This might be explained by a shift in the vegetative cycle, as flowering should have occurred earlier in coastal regions than in inland elevated areas.

The sensitivity of the contamination of grapes and wine to the vegetative stage at the time of deposition has been studied by Levain *et al.* (2006) using a more theoretical approach. This approach was based on the work of Madoz-Escande *et al.* (1997), Carini *et al.* (1996), and Carini and Lombi (1997), on the foliar interception equations proposed in ECOSYS (Müller and Pröhl, 1993) and using an agronomic model to simulate the growth of various kinds of vines. For a wet deposition which occurred 4 to 6 months before the harvest (end of March to June), T_{ag} values calculated by Levain *et al.* (2006) would range from 2×10^{-4} to 4×10^{-3} $m^2 \cdot kg^{-1}$ fw depending on the amount of rainfall during deposits. However, the values become close to zero for a longer period (>6 months) before harvest (in March notably) because the vines are assumed to be at too early a stage to have intercepted deposits. For a wet deposit which occurred around 1 month before harvest, the theoretical T_{ag} value would range between 9×10^{-3} $m^2 \cdot kg^{-1}$ (amount of rain of 15 mm) and 2×10^{-2} $m^2 \cdot kg^{-1}$ (amount of rain of 5 mm). The observations made for sour cherries sampled in northeastern France in 1986 match the later theoretical scenario well. Sour cherries were sampled from mid-June to the beginning of July (*i.e.* 1.5 to 2 months after deposition).

3 Annual decrease in fruit contamination in Fukushima Prefecture

For the six fruits considered, Table 1 provides massic activities for the harvests of 2012 and 2013. Like in 2011, the most contaminated fruits are the earliest species that are produced in the most contaminated areas: apricots from Kori-machi, Fukushima-shi and Date-shi municipalities. Compared with 2011, activities and T_{ag} values (although not shown in Tab. 1) decreased by an overall factor of 5 in 2012 and 10 in 2013. Only the fruit species with the earliest development still had detectable levels of contamination in 2013, *i.e.* apricot and cherries.

Based on our current knowledge of element cycling in orchard and forest trees (Antonopoulos-Domis *et al.*, 1991, 1996; IAEA, 2003; Goor and Thiry, 2004; Brown and Sherwood, 2012), this lasting contamination in tree fruits some years after the accident is partly caused by the internal transfer of the initially intercepted caesium. Part of the caesium which has been intercepted by the aerial parts in March 2011, *i.e.* apricot tree flowers or buds and twigs for other species, must

have been incorporated into internal tissues and translocated to other tree organs. Once incorporated, caesium is basically translocated either to the growing fruits during their maturation or into the stem and wood storage compartments at the predormancy stage in fall. Other potential sources of contamination of fruit in subsequent years are the re-deposition on leaves and fruits of caesium resuspended in the air from the soil surface or leached during rain from tree bark initially exposed to radioactive fallout (Takata, 2013). It is effectively well known that the contribution of root uptake from contaminated soil is very limited during the first few years after an accident. For example, field sampling by Antonopoulos-Domis *et al.* (1996) in Greek orchards showed that, in their case, the root pathway contribution was not detectable at least during the first 5 years following the Chernobyl fallouts. This is actually due to the fact that caesium migration into the underlying soil layers is very slow and that the soil-to-fruit transfer factor is also quite low. Characteristic values of transfer factors for caesium can be found in the literature (Müller and Pröhl, 1993; Carini, 2001; Brown and Sherwood, 2012). They typically vary between 5×10^{-3} and 2×10^{-2} $kg \text{ dw soil} \cdot kg^{-1}$ fw fruit. If we assume that tree roots are mainly confined in the top 30-cm soil layer and that caesium is already homogeneously distributed in this layer (which is a very pessimistic assumption), a 100 $kBq \cdot m^{-2}$ surface activity would have led to only a few $Bq \cdot kg^{-1}$ fw fruit in 2012.

Figure 2 shows the decrease in the T_{ag} values ($m^2 \cdot kg^{-1}$ fw) for all kinds of fruits. Like in 2011, T_{ag} values remain quite variable in 2012 and 2013, and may vary by one order of magnitude (for apricots). We presume that this variability is directly inherited from the variability in 2011, as discussed in the previous paragraph. Effective half-lives have been calculated for both years and each type of fruit, grouping all municipalities together, which are not statistically different. Effective half-lives for 2011–2012 range from 172 d for apricots to 264 d for pears, with a mean value of about 200 d. Although more uncertain, the estimates for 2012–2013 seem to be longer than the previous ones, at least for apricots (much uncertainty exists in the estimate for cherries because data are too scarce in 2013).

Similar observations were made by Mück (1997) on the basis of Austrian apples and pears sampled from 1986 to 1993. The effective half-life increased from 91 d for pears and 98 d for apples in 1986–1987, to 450 d and 507 d, respectively, from 1987 to 1993. Antonopoulos-Domis and Clouvas (1991) also estimated half-lives for cherries, apricots, pears and apples from their sites in Greece, but for the whole 1987–1990 period, without distinguishing years. Their estimates ranged from 241 d to 314 d with a mean value of about 270 d. This is also the case of global half-lives deduced from the data originating from Date and Fukushima municipalities. In these two areas, no measurements were available in 2012, but the global effective half-lives for the period 2011–2013 comprised between 186 d and 272 d, respectively. These pluri-annual half-lives are logically intermediate between those corresponding to the first-to-second year ones and those of following years obtained after the Chernobyl accident, in Austria and Italy (188 d for the period 1986–1988 from Monte *et al.* 1990).

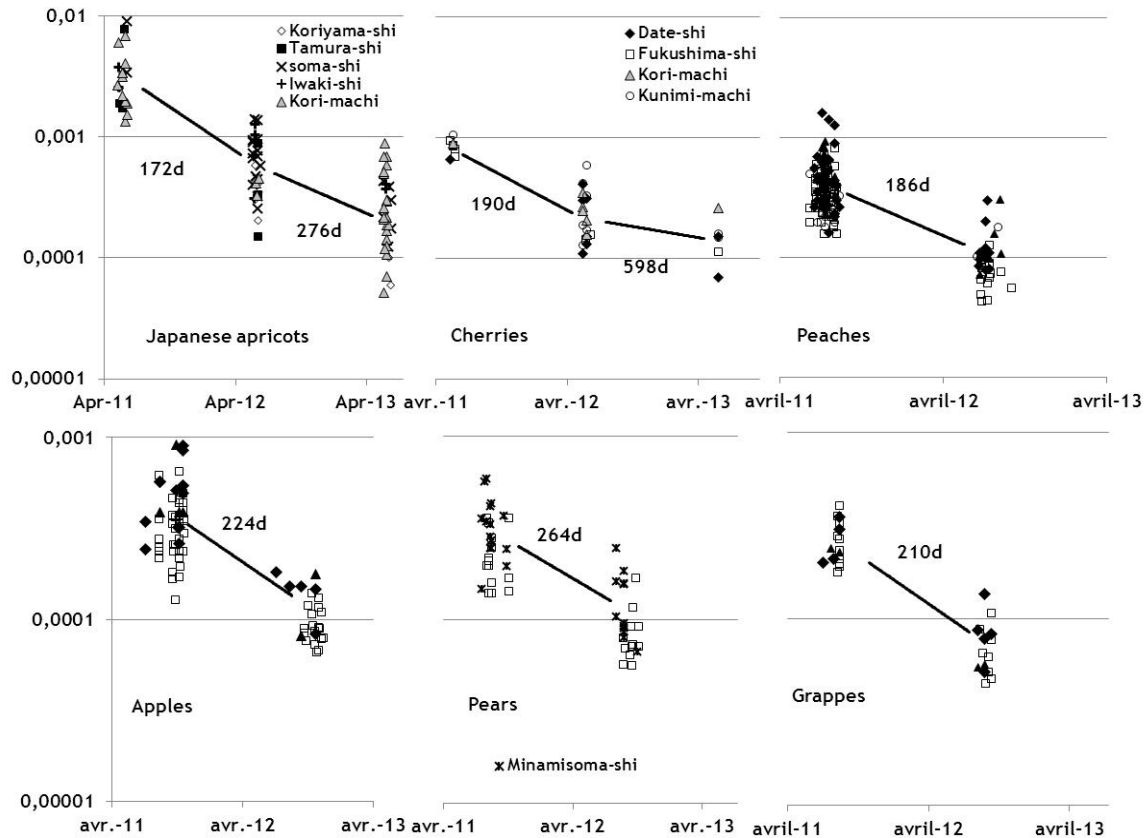


Fig. 2. Effective half-lives (in days) of contamination in orchard fruits from Fukushima Prefecture.

4 Conclusion

Data on fresh fruits collected after the Fukushima and Chernobyl accidents show that for radioactive fallouts occurring in the middle of spring (the case of Chernobyl) or in late winter (the case of Fukushima), the contamination of fruit, although lower than in cases where accidents had occurred in summertime, remains significant and persistent during the following years. The interception of airborne contamination, even in the absence of leaves (the case of Fukushima), leads to a significant potential of lasting fruit contamination for the following years. The observations made in 1986 in France, Italy, Greece and Austria, and Japan in 2011, show that the growth stage of tree fruit at the time of deposition is a very sensitive factor. Our meta-analysis enabled us to estimate the change in aggregated transfer factor values for radiocaesium, expressed in $\text{m}^2 \cdot \text{kg}^{-1} \text{fw}$ (T_{ag}), as a function of the deposition-to-harvest delay. For a predominant wet deposition, T_{ag} values decrease from a few $10^{-2} \text{m}^2 \cdot \text{kg}^{-1} \text{fw}$ to a few 10^{-4} when the time elapsed between fallout and fruit harvest increases from about a month to seven months. Notably, the value of $2 \times 10^{-4} \text{m}^2 \cdot \text{kg}^{-1}$ seems to be the lowest which can be attributed to this aggregated transfer factor, even for wet deposits occurring in winter.

The deposition-to-harvest delay, which is the usual input data for post-accidental food-chain transfer models, only partly accounts for the fruit growth stage at the time of deposition. As discussed before, the transfer to fruits is so sensitive at some key development stages that the aggregated transfer

factor can vary by more than a factor of 3 to 5 under the influence of specific soil and climate conditions. The prediction of fruit contamination by a model based on the deposition-to-harvest delay would be more accurate if key development dates, such as budding or flowering, were precisely known, site-specific and adapted to the year of accidental deposits.

Recent Japanese data are in good agreement with the few observations made after the Chernobyl accident. They show that the contamination of fruits decreases with an effective half-life of about 170 d to 260 d from the first to the second year after radioactive fallout. Variations in half-lives obviously depend on the growth stage at the time of deposition. From the second to the third year after deposit, half-lives are longer and vary between 300 and 600 d.

References

- Anguissola Scotti I., Silva S. (1992) Foliar absorption and leaf-fruit transfer of ^{137}Cs in fruit trees, *J. Environ. Radioact.* **16**, 97-108.
- Antonopoulos-Domis M., Clouvas A., Gagianas A. (1991) Radiocaesium dynamics in fruit trees following the Chernobyl accident, *Health Phys.* **61**, 837-842.
- Antonopoulos-Domis M., Clouvas A., Gagianas A. (1996) Long-term radiocaesium contamination of fruit trees following the Chernobyl accident, *Health Phys.* **71** (6), 910-914.
- Brown J., Simmonds J. (1995) FARMLAND: a dynamic model for the transfer of radionuclides through terrestrial foodchains, Report NRPB-R273. Chilton, UK.

- Brown J., Sherwood J. (2012) Modelling approach for the transfer of radionuclides to fruit species of importance in UK. Health Protection Agency, Centre for Radiation, Chemical and Environmental Hazards Chilton, Didcot, Oxfordshire OX11 0RQ.
- Carini F. (1999) Radionuclides in plants bearing fruit: an overview, *J. Environ. Radioact.* **46**, 77-97.
- Carini F. (2001) Radionuclide transfer from soil to fruit, *J. Environ. Radioact.* **52**, 237-279.
- Carini F., Anguissola Scotti I., Montrucoli M., Silva S. (1996) ^{134}Cs foliar contamination of vine: translocation to grapes and transfer to wine. In: *Proceedings of the International Symposium on Radioecology: ten years terrestrial radioecological research following the Chernobyl accident*, Austrian Soil Science Society and Federal Environment Agency, Vienna, Austria (M. Gerzabek, Ed.) pp. 163-169.
- Carini F., Lombi E. (1997) Foliar and soil uptake of ^{134}Cs and ^{85}Sr by grape vines, *Sci. Total Environ.* **207**, 157-164.
- Carini F., Bengtsson G. (2001) Post-deposition transport of radionuclides in fruits, *J. Environ. Radioact.* **52**, 215-236.
- Champion D., Korsakissok I., Didier D., Mathieu A., Quelo D., Groell J., Quentric E., Tombette M., Benoit J.P., Saunier O., Parache V., Simon-Cornu M., Gonze M.A., Renaud Ph., Cessac B., Navarro E., Servant-Perrier A.C. (2013) The IRSN's earliest assessments of the Fukushima accident's consequences for the terrestrial environment in Japan, *Radioprotection* **48** (1), 11-37.
- Goor F., Thiry Y. (2004) Processes, dynamics and modelling of radio-caesium cycling in a chronosequence of Chernobyl-contaminated Scots pine (*Pinus sylvestris* L.) plantations, *Sci. Total Environ.* **325**, 163-180.
- International Atomic Energy Agency (2003) Modelling the transfer of radionuclides to fruit. Report of the fruits working group of BIOMASS theme 3. IAEA, Vienna. ISBN 92-0-106503-5.
- Katata G., Terada H., Nagai H., Chino M. (2012a) Numerical reconstruction of high dose rate zones due to the Fukushima Daiichi Nuclear Power Plant accident, *J. Environ. Radioact.* **111**, 2-12.
- Katata G., Ota M., Terada H., Chino M., Nagai H. (2012b) Atmospheric discharge and dispersion of radionuclides during the Fukushima Daiichi Nuclear Power Plant accident. Part I: Source term estimation and local-scale atmospheric dispersion in early phase of the accident, *J. Environ. Radioact.* **109**, 103-113.
- Korsakissok I., Mathieu A., Didier D. (2013) Atmospheric dispersion and ground deposition induced by the Fukushima Nuclear power plant accident: a local-scale simulation and sensitivity study, *Atmos. Environ.* **70**, 267-279.
- Levain A., Mercat-Rommens C., Roussel-Debet S. (2006) Study of the radioecological sensitivity of vine after a radioactive pollution. Report IRSN/DEI/SESURE-2006-54 (in French).
- Madoz-Escande C., Colle C., Hugon J., Jouglet H., Tormos J. (1997) Foliar contamination of vine crops. 1994-1995 lysimeter experiments on large soil samples extracts of the Tricastin site. Report IPSN/DPRE/SERE 97/026 (in French).
- Madoz-Escande C., Colle C., Brechignac F., Jouglet H., Fernandez V. (1998) Time dependent evolution of ^{137}Cs and ^{90}Sr deposited as aerosols on vine. Poster presented at the 8th annual meeting of SETAC-Europe, Bordeaux 14-18 April 1998.
- Maubert H., Roussel S. (1988) Results and interpretations of observations from the South-eastern France after the Chernobyl radioactive deposits in May 1986. Report CEA/IPSN/SERE, June 1988 (in French).
- MEXT (2013) Monitoring information of environmental radioactivity level, <http://radioactivity.nsr.go.jp/en/>
- MHLW (2013) Information on the Great East Japan Earthquake, <http://www.mhlw.go.jp/english/topics/2011eq/index.html>
- Monte L., Quaggia S., Pompei F., Fratarcangeli S. (1990) The behavior of ^{137}Cs in some edible fruits, *J. Environ. Radioact.* **11**, 207-214.
- Mück K. (1997) Long-term effective decrease of caesium concentration in foodstuffs after nuclear fallout, *Health Phys.* **72** (5), 659-673.
- Müller H., Pröhl G. (1993) ECOSYS-87: a dynamic model for assessing radiological consequences of nuclear accidents, *Health Phys.* **64** (3), 232-252.
- Pröhl G. (2009) Interception of dry and wet deposited radionuclides by vegetation, *J. Environ. Radioact.* **100**, 675-682.
- Renaud Ph., Maubert H., Bernié J.C. (1997) The contextual parameters in post-accidental radioecology, *Radioprotection* **32** (2), 181-195 (in French).
- Renaud Ph., Pourcelot L., Métivier J.M., Morello M. (2003a) Mapping of ^{137}Cs deposition over eastern France 16 years after the Chernobyl accident, *Sci. Total Environ.* **309**, 357-364.
- Renaud Ph., Colle C., Louvat D., Barker E. (2003b) Influence des retombées de l'accident de Tchernobyl sur l'activité en ^{137}Cs du vin des côtes du Rhône, *Radioprotection* **38** (1), 51-59.
- SCPRI (1986) Bulletins mensuels de mesures du Service Central de protection contre les Rayonnements ionisants. Juillet 1986.
- Silva S., Anguissola Scotti I., Fregoni M., Vercesi A. (1989) Contamination radioactive du raisin en Italie, en 1986, à la suite de l'accident de la centrale nucléaire de Tchernobyl, *Bulletin de l'OIV* **62** (695-696), 74-84.
- Takata D. (2013) Distribution of Radiocaesium from the Radioactive Fallout in fruit trees. In: *Agricultural Implications of the Fukushima Nuclear Accident* (Nakanishi and K. Tanoi, Eds.) Chap. 14, pp.143-162.
- Terada H., Katata G., Chino M., Nagai H. (2012) Atmospheric discharge and dispersion of radionuclides during the Fukushima Daiichi Nuclear Power Plant accident. Part II: verification of the source term and analysis of regional-scale atmospheric dispersion, *J. Environ. Radioact.* **112**, 141-154.