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Transfer of ^{131}I from Fukushima to the Vegetation and Milk in France

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ABSTRACT: Iodine-131 and various other radionuclides were released into the atmosphere from the damaged Japanese reactors of Fukushima Dai-ichi from 12 to 22 March 2011. The contaminated air mass was detected in France after 24 March; samples of grass, vegetables, and milk have been analyzed for ^{131}I by the IRSN, considering the fact that few values of iodine-131 transfer parameters have been directly measured *in situ*, due to the radioactive decay of this isotope. Data are compared with calculated values according to the air iodine concentration. The apparent dry deposition velocity of iodine on grass is therefore estimated to range between 1×10^{-3} and 5×10^{-3} m s^{-1} from site to site. In addition, the grass to milk transfer factors are 2.8×10^{-2} and 3.6×10^{-3} d L^{-1} for goat's and cow's milk respectively. These parameters fit well with the current values usually considered for radioecological assessment.

1. INTRODUCTION

The major release of radioactive materials into the atmosphere from the reactors accident occurring at the Fukushima Dai-ichi nuclear power station (Japan) lasted nearly 10 days, from 12 to 22 March 2011.[1] During this period the release of iodine-131 was estimated at 2×10^{17} Bq by the Nuclear and Industrial Safety Agency (NISA) and the Institute of Radioprotection and Nuclear Safety (IRSN). Modeling the atmospheric dispersion of the radioactive releases and measurements carried out in the air have shown that the radioactive plume, involving mainly gaseous and volatile radionuclides such as noble gases, tellurium, cesium, and iodine isotopes, drifted over the Pacific Ocean, North America, and the Atlantic Ocean, before reaching North European countries.[10] The radioactive plume was expected after 23 March in France (CMRS Toulouse, Météo France). The assessment of radionuclides contamination in vegetation after a nuclear atmospheric release is of significant importance because contamination of vegetable products allows the evaluation of the transfer to the food chain. For this reason measurements of ^{131}I (half-life = 8.05 days), tracer of this accidental release, have been carried out in the air, in vegetation and milk between 21 March and 10 May 2011 by IRSN, at various sites located in the south of France.

Whereas the intensity of the depositions of radioactive particles on the ground depends widely on the intensity of precipitation,[2−4] the ^{131}I activity in the vegetation is closely related to the activity in the air, since rain washing of radioactive particles on the aerial biomass is often assumed.[5] The empirical equations used to estimate the activity in vegetation varies from one model to another, taking into account various parameters such as the concentration in the air; the duration of deposit; the amount of biomass; the decay over time, due to both radioactive decay and dilution due to the growth of the plant; the deposition velocity. This latter parameter is estimated using different approaches. Thus, apparent deposition velocity of iodine of 3×10^{-3} m s^{-1} was derived from measurements following the Chernobyl accident carried out in the air during a period of time and for a unit surface of vegetation.[6] More recent experimental work on dry deposition of gaseous ^{131}I underlines the significantly higher iodine deposition (10^{-3} m s^{-1} in grass, deduced from the concentration in grass and the flux of iodine in the greenhouse) coarsely 1 order of magnitude higher than that of particulate radionuclides such as radiocesium.[2] Lastly, typical default values given for the modeling of dry deposits on grass are 1.5×10^{-2} and 1.5×10^{-3} m s^{-1} for gaseous and particulate iodine, respectively.[7] The ECOSYS model assumes deposition velocity is widely dependent on the stage of development of the plant canopy, characterized by leaf area index (LAI), strongly dependent on the time of year.

Whereas dry deposition velocity proposed soon after Chernobyl was estimated from the integrated activity measured in the air and the whole activity deposited on vegetation after the Chernobyl releases, the *in situ* time trends of the iodine activity reported in the present paper allow us to propose transfer parameters for iodine as can be derived from observations made in France after the Fukushima accident. In this respect the activity measured in vegetation according to time at four different sites is compared with the activity in the air and fitted values of the apparent velocity of iodine are deduced thereafter. Furthermore the present field data have also allowed evaluating complementary values of the *in situ* transfer factors for ^{131}I to goat's milk, which was derived from scarce results up to now.[8] Thus, the aim

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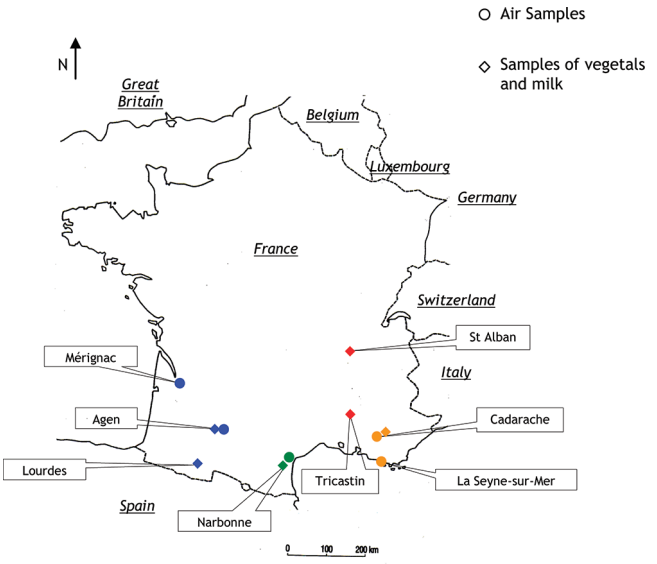

Figure 1. Location of vegetation, milk, and air sampling sites.

of this study, derived from the field work, is to better constrain the transfer of radioiodine to plants and milk in case of accidental release.

2. SAMPLING AND MEASUREMENTS

Activity of ^{131}I was acquired simultaneously in three compartments in the terrestrial environment including the air (sampling frequency: 2−3 days), the vegetation (every 2−3 days), and the milk (every 3 days). The sampling effort was focused on four sites located in the SE (Cadarache and Tricastin) and SW of France (Narbonne and Agen) (Figure 1). In addition, the air activity was also measured at two other sites located in SW (Mérignac) and SE (La Seyne-sur-Mer), as close as possible to the sites of Agen and Cadarache, respectively.

Between 24 March and the beginning of May 2011, atmospheric particles supporting ^{131}I have been collected on filters Jonell ref JPME 13 160 (surface 1316 cm^2 and minimum collection efficiency of 94.5% for 30 nm particle sizes) using high-volume samplers having a flow rate of 300−600 m^3 h^{-1} of the OPERA network monitoring radioactivity in the air in the SE (Cadarache and La Seyne-sur-Mer) and SW of France (Narbonne and Mérignac).[9] For trapping gaseous iodine a charcoal filter was put underneath the aerosol filter (3−10 m^3 h^{-1}) (Agen and Cadarache). As aerosol and gaseous iodine fractions were not both available at the same site/period, and it needed to be evaluated from gaseous iodine/total iodine ratio (see section 3.2).

In addition approximately three or more kilograms of grass and leaf vegetables (lettuce or spinach) have also been collected in the surrounding fields in Agen, Lourdes (located some 130 km from Agen), Narbonne, Cadarache, Tricastin, and in St-Alban (located some 100 and 220 km respectively from Tricastin and Cadarache in the Rhône valley). At three sites (namely Cadarache, Tricastin, and Agen) grass covering one meter square was collected, allowing us to evaluate the yield of grass (Y in kg fresh weight m^{-2}, see eq 2). At the same time goat's milk (Cadarache, Tricastin, and St-Alban) and cow's milk (Agen) were also sampled, when herds were grazing in the fields. Samples were immediately transferred in 3 L Marinelli beakers. ^{131}I activity has been determined using very low background (filters and rain waters) and low background (others samples) Ge-gamma spectrometers, based on main peak occurring at 364.5 keV (81%). Although the determination of ^{131}I occurred less than 24 h after sampling, the activity of ^{131}I was decay-corrected to the sampling date.

3. RESULTS AND DISCUSSION

3.1. Activity Measured in Vegetation and in the Air. Due to the absence of iodine-131, below detection limit values are observed before 28 March at all sites; thereafter an increase of iodine is observed (Table 1). The highest values are reached at SW sites (for example: 9.0 Bq kg^{-1} fresh weight at Agen or 3.6 Bq kg^{-1} fresh weight at Lourdes), whereas SE sites record lower values, close to 1 Bq kg^{-1} fresh weight roughly. Thereafter the activity slowly decreases and reaches detection limit (<0.1 Bq kg^{-1} fresh weight) by 10 May. The leaves of vegetables exhibit lower activity than grass taken at the same site/date; however, our set of data does not evidence any higher activity occurring in samples taken at SW sites (Table 1).

The whole set of activity measured in the air is detailed elswhere.[10] The main variations recorded in the air are shortly recalled thereafter (Figure 2). As noticed before for grass, highest iodine activity in the air is measured at the SW sites especially in Agen (9.6 mBq m^{-3} on 25−27 March) and in Mérignac (3.2 mBq m^{-3} on 24−27 March). Maximum activity occurred later in Narbonne (2.5 mBq m^{-3} on 28−30 March). Thereafter the activity slowly decreases with time as shown by Agen data, due to the rain which lasted for five days (26−30 March). During sampling time, the precipitations vary from site to site, between 4.4 mm (in Agen) and 34 mm (in Cadarache). Although it is not used to evaluate the deposition on vegetation, rain episodes should induce a decrease of the activity in the air. However, the activity measured in the air remains quite constant in Mérignac, probably due to the arrival of contaminated air masses after rain. Set against this, at the SE sites, activity never exceeds 4 mBq m^{-3} (Cadarache, 30 March) and 1.4 mBq m^{-3} (La Seyne-sur-Mer, 28−30 March), soon after rainy days (27−30 March). Again the arrival of contaminated air masses after rainfall is assumed. Thus air activity data highlight chronic contamination in the atmosphere lasted about 8−10 days, with higher activity at SW sampling sites compared to SE ones.

The comparison of the time trends of activity in the vegetation and in the air sampled at the same site highlights similar pattern, despite a time lag of 1−2 days between both compartments. Also rather smooth variations are noticeable in the vegetation with respect to the atmospheric compartment. For example, the activity peaks in the air on 30 March in Cadarache and the level in vegetation increases by a factor of 3, two days later. Similarly, maximum activity is reached in the vegetation taken in Narbonne two days later than in the air. Due to lack of data between 29 March and 4 April in Tricastin we probably missed the maximum activity in grass expected during this period, according to the air's trend peaking on 30 March. Lastly a conjunction of strongest activities in the vegetation and in the air at the SW sites (Agen) is noticed. A similar pattern of the activity observed in the air and in the vegetation is mainly accounted for by dry deposition prevailing for iodine.[11]

3.2. Apparent Deposition Velocity of Iodine-131. A preliminary estimation of the gaseous iodine (not available at all sampling points) was necessary. It was considered that gaseous iodine is 73 ± 15% of the total iodine, according to measurements

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Table 1. ^{131}I Activity Measured in Vegetation

sampling site species	date	(Bq kg^{-1} fresh weight)	sampling site species	date	(Bq kg^{-1} fresh weight)
Agen	21/03/2011	<0.1	**Tricastin**	29/03/2011	0.3 ± 0.1
grass	25/03/2011	<0.3	spinach/lettuce	04/04/2011	0.3 ± 0.1
	28/03/2011	3.6 ± 0.4		04/04/2011	0.4 ± 0.1
	30/03/2011	9 ± 1		06/04/2011	0.4 ± 0.1
	01/04/2011	3.97 ± 0.03		08/04/2011	0.6 ± 0.1
	04/04/2011	2.0 ± 0.2		11/04/2011	0.4 ± 0.1
	06/04/2011	2.1 ± 0.4		14/04/2011	0.21 ± 0.09
	08/04/2011	1.2 ± 0.2		19/04/2011	0.12 ± 0.04
	12/04/2011	0.7 ± 0.2	**Narbonne**	22/03/2011	<0.1
	15/04/2011	0.8 ± 0.1	grass	28/03/2011	0.7 ± 0.1
	20/04/2011	0.4 ± 0.1		30/03/2011	2.3 ± 0.3
	28/04/2011	0.13 ± 0.07		01/04/2011	2.8 ± 0.4
Cadarache	22/03/2011	<0.1		04/04/2011	2.9 ± 0.5
lettuce	25/03/2011	<0.1		07/04/2011	1.6 ± 0.2
	28/03/2011	<0.1		14/04/2011	1.1 ± 0.2
	30/03/2011	0.12 ± 0.04		20/04/2011	0.9 ± 0.2
	01/04/2011	0.08 ± 0.03		27/04/2011	0.2 ± 0.1
	04/04/2011	0.09 ± 0.05	**Narbonne**	22/03/2011	<0.1
Cadarache	21/03/2011	<0.2	spinach	24/03/2011	<0.1
grass	25/03/2011	<0.1		28/03/2011	0.14 ± 0.06
	28/03/2011	0.6 ± 0.2		30/03/2011	1.0 ± 0.1
	30/03/2011	0.7 ± 0.1		01/04/2011	0.7 ± 0.1
	01/04/2011	1.3 ± 0.1	**St-Alban**	29/03/2011	0.5 ± 0.1
	04/04/2011	0.9 ± 0.2	grass	01/04/2011	0.7 ± 0.2
	06/04/2011	0.9 ± 0.2		04/04/2011	1.0 ± 0.2
	08/04/2011	0.4 ± 0.1		11/04/2011	1.0 ± 0.2
	11/04/2011	0.4 ± 0.1		19/04/2011	0.5 ± 0.1
	13/04/2011	0.3 ± 0.1	**St-Alban**	23/03/2011	<0.1
	18/04/2011	0.3 ± 0.1	lettuce/spinach	26/03/2011	<0.1
	27/04/2011	<0.1		29/03/2011	0.2 ± 0.1
Tricastin	29/03/2011	2.7 ± 0.3		01/04/2011	0.5 ± 0.1
grass	04/04/2011	1.4 ± 0.2		04/04/2011	1.2 ± 0.2
	06/04/2011	1.5 ± 0.3	**Lourdes**	28/03/2011	2.0 ± 0.3
	08/04/2011	1.3 ± 0.1	grass	31/03/2011	3.6 ± 0.5
	11/04/2011	1.5 ± 0.3		04/04/2011	2.0 ± 0.4
	14/04/2011	1.2 ± 0.2			
	19/04/2011	0.4 ± 0.1			
	28/04/2011	0.2 ± 0.1			
	03/05/2011	0.12 ± 0.05			
	10/05/2011	<0.08			

carried out by European scientists belonging to the Ring of Five, which is an informal group for the purpose of exchanging data on concentration of radionuclides in the air.[10]

The deposition of iodine released from Fukushima reactors was computed as a daily accumulation on the ground vegetation. The input is estimated based on iodine measurements carried out in the air (total iodine involving particulate+gaseous fractions), detailed in the previous section.

The daily activity in the air allows us to estimate dry deposition occurring every day, by using an apparent deposition velocity

$$D(d) = C_{air}(d) \times Vd_{app} \quad (1)$$

where d is the considered day (date); $D(d)$ is the deposited iodine on the day d (Bq m^{-2}); C_{air} is the total iodine activity in the air (Bq m^{-3}); and Vd_{app} is the apparent deposition velocity (m s^{-1}) adjusted so that the calculated activity in vegetation (calculated with eqs 2 and 3 thereafter) fit with activity measured in vegetal samples.

The root transfer is insignificant here; therefore, we only take into account the foliar transfer. From the comparison of air and vegetation data (see previous section), it appears that dry deposition was significant for iodine, and thus the contamination of grass and leaves of vegetables was computed as dry deposition of iodine, following eqs 2 and 3 respectively, derived from ASTRAL model[12,13]

$$C_{grass}(d) = \frac{Rc}{Y} \times \sum_{0}^{d} D(d) \times \exp -[(\lambda_{wb} + \lambda_r) \times \Delta t] \quad (2)$$

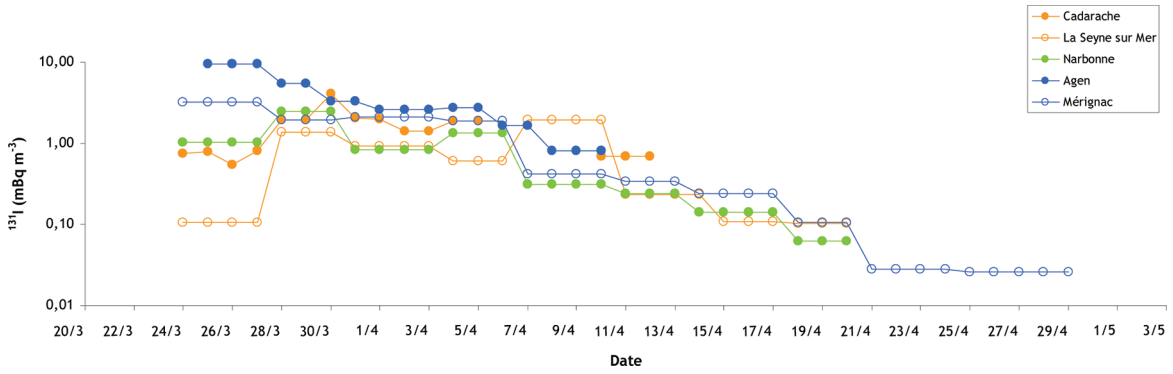

Figure 2. ^{131}I activity measured in the air according to time.

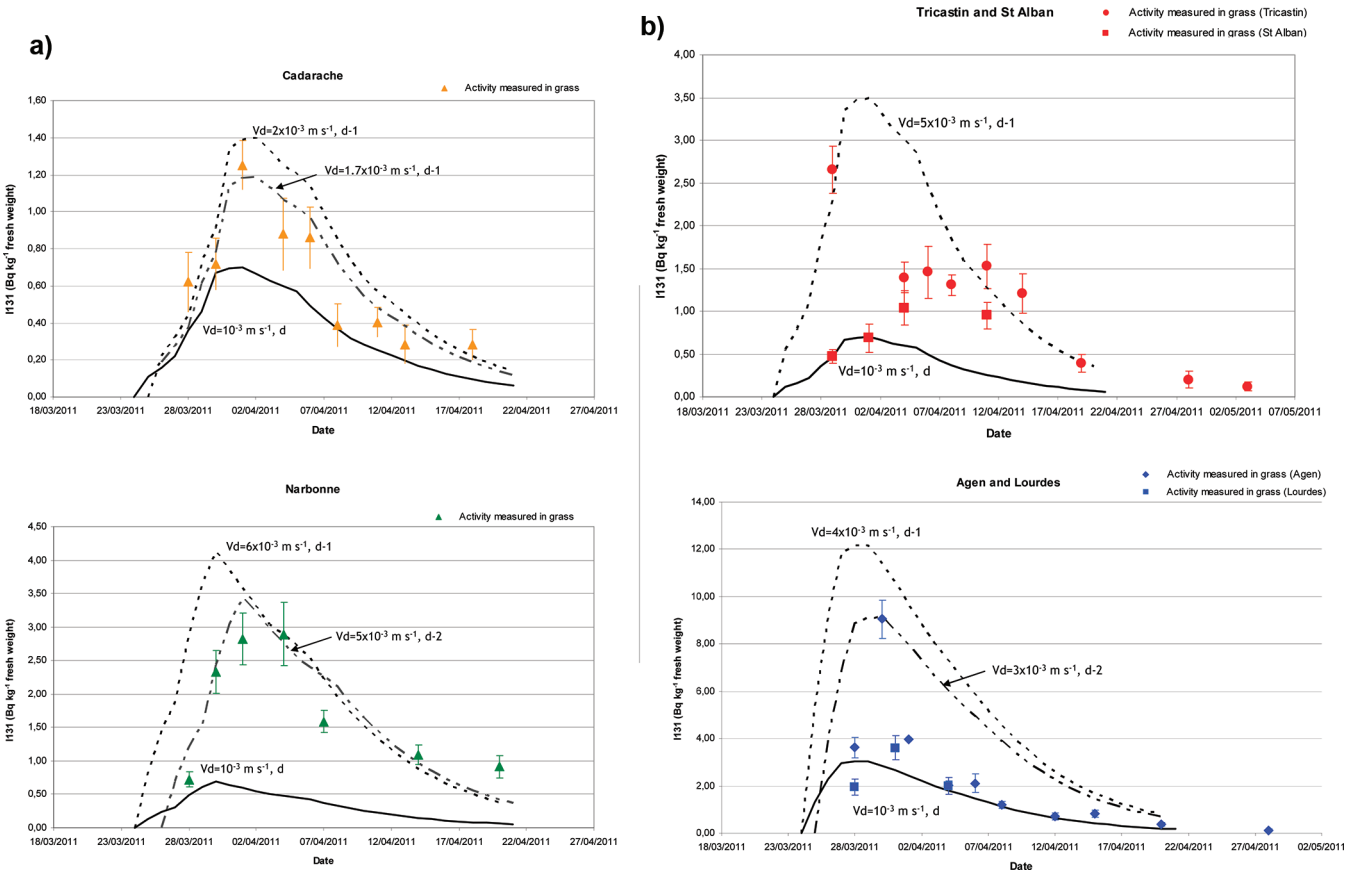

Figure 3. ^{131}I activity in grass deduced from apparent dry deposition velocity compared with ^{131}I activity measured in grass.

$$C_{veg}(d) = \sum_{0}^{d} D(d) \times Ft \times \exp -[(\lambda_{wb} + \lambda_r) \times \Delta t] \quad (3)$$

where $C_{grass}(d)$ or $C_{veg}(d)$ is the grass or vegetable measured concentration due to the deposit of the considered day d (Bq kg^{-1} fresh weight); $D(d)$ is the deposited iodine on day d (Bq m^{-2}); Y and R_c are the yield (kg fresh weight m^{-2}) of grass and the retention ratio for deposit on grass, respectively. R_c/Y ratio varies linearly between 1.4 (March) to 0.56 m^2 kg^{-1} fresh weight (May); the value Y used in ASTRAL (0.7 kg fresh weight m^{-2}) is quite close to the yield given by field: 1.65, 0.48, and 1.18 kg fresh weight m^{-2} in Cadarache, Tricastin, and Agen, respectively; Ft is the dry foliar transfer factor of iodine for leafy vegetables (m^2 kg^{-1} fresh weight), equal to 5×10^{-1} m^2 kg^{-1}; λ_{wb} is the loss rate due to weathering and biomass growing (d^{-1}); λ_{wb} is considered equal to 1×10^{-1} d^{-1} for vegetables and 6×10^{-2} d^{-1} for grass; λ_r is the radioactive decay rate (d^{-1}) equal to 8.61×10^{-2} d^{-1}; and Δt is the time span between the beginning of deposition and the day of sampling (d).

Other parameters apart from deposition velocity were considered as default values in the equations. Indeed default parameters of ASTRAL fit well with values given by the IAEA EMRAS working group which aim to validate the ^{131}I ecological transfer models: the yield of grass ranging between 0.1 and 0.6 (kg fresh weight m^{-2}) and the R_c/Y ratio ranging between 0.5 to 1.65.[14] Also λ_{wb} was chosen between 0.05 and 0.1 d^{-1} by this working group. In comparison with those "consensual" parameters,

Table 2. ^{131}I Activity Measured in Milk

sampling site species	date	(Bq L^{-1})	sampling site species	date	(Bq L^{-1})
Agen	21/03/2011	<0.061	**Cadarache**	21/03/2011	<0.062
cow's milk	28/03/2011	<0.108	goat's milk	25/03/2011	<0.050
	30/03/2011	0.455 ± 0.121		28/03/2011	<0.068
	01/04/2011	0.473 ± 0.073		30/03/2011	<0.141
	04/04/2011	0.448 ± 0.066		01/04/2011	1.026 ± 0.060
	06/04/2011	0.659 ± 0.088		04/04/2011	0.787 ± 0.078
	08/04/2011	0.329 ± 0.068		06/04/2011	0.304 ± 0.053
	12/04/2011	0.139 ± 0.064		08/04/2011	0.686 ± 0.063
	15/04/2011	0.104 ± 0.048		11/04/2011	0.348 ± 0.087
	20/04/2011	0.045 ± 0.031		13/04/2011	0.193 ± 0.210
	11/05/2011	<0.043		18/04/2011	0.116 ± 0.057
Tricastin	29/03/2011	0.993 ± 0.175		27/04/2011	<0.070
goat's milk	01/04/2011	2.120 ± 0.219	**St-Alban**	23/03/2011	<0.062
	04/04/2011	1.210 ± 0.177	goat's milk	26/03/2011	<0.061
	06/04/2011	1.691 ± 0.178		29/03/2011	<0.171
	08/04/2011	1.536 ± 0.150		01/04/2011	<0.076
	11/04/2011	0.650 ± 0.084		04/04/2011	0.460 ± 0.089
	14/04/2011	0.617 ± 0.121		12/04/2011	0.18 ± 0.044
	18/04/2011	0.410 ± 0.056		19/04/2011	0.280 ± 0.049
	28/04/2011	0.076 ± 0.023			
	03/05/2011	0.061 ± 0.021			
	10/05/2011	<0.043			

Table 3. Transfer Coefficient of Iodine to Goat's and Cow's Milk (d L^{-1})[a]

	goat's milk				cow's milk			
			min				min	
source	AM	SD	max	n	AM	SD	max	n
this study	0.28	0.15	0.06	14	3.6×10^{-3}	6.2×10^{-4}	1.0×10^{-3}	8
			0.6				6.2×10^{-3}	
Howard et al., 2009	0.33	0.23	0.027	24	9.1×10^{-3}	7.0×10^{-3}	4.0×10^{-4}	104
			0.77				2.5×10^{-2}	

[a] AM: arithmetic mean; SD: arithmetic standard deviation; n: number of values of the transfer coefficient.

the velocity of iodine was highly variable (two to 3 orders of magnitude) from one model to another and also depending on the speciation in the air (particulate, gaseous, organic phases).[14]

In the present study the activity in grass was computed for different apparent deposition velocity values and compared to that measured in the grass sampled at each studied site (Figure 3). Computation also takes into account the time lag between the atmospheric compartment and ground vegetation reported in the previous section. The origins of the curves were adjusted when necessary, beginning at (*d-1*) or (*d-2*), to better fit the grass activity.

The computed evolution of the activity in grass with respect to time fit roughly with the field data, with an increase followed by a decrease in the activity (Figure 3). Despite such large variation in grass rather homogeneous deposition velocity is deduced (from 1×10^{-3} to 5×10^{-3} m s^{-1}). Thus the highest activities noticed in the SW would have been induced by slightly higher deposition rates (5×10^{-3} m s^{-1} in Narbonne, for example) in conjunction with enhanced activity in the air (see section 3.1) (Figure 3a). In the Rhône valley also slightly higher velocity is assumed in Tricastin (3×10^{-3} m s^{-1}) with respect to St-Alban (10^{-3} m s^{-1}) (Figure 3b). In addition to site to site, variation of deposition velocity should occur naturally with time. Indeed at a single site such as Agen, the highest activity (9.0 Bq kg^{-1}) is consistent with higher velocity (3×10^{-3} m s^{-1}) compared with the rest of the data set which yields 1×10^{-3} m s^{-1} (Figure 3b). In another case (Narbonne), an improvement of the fitting of computed/field data is obtained when activity measured in the air two days before are taken into account (Figure 3a).

Various assumptions are usually made about the interception of airborne and waterborne radionuclides by vegetation, especially since the fraction of dry and wet deposition is unknown and the interception factors of both remain controversial.[14] The present work does not take into account the wet deposition of iodine, although a part of the iodine measured in vegetation (even if it is low) is deposited with rain (4.4 to 34 mm from site to site during the sampling period). Thus apparent velocities determined by fitting with the field activity of vegetation overestimate

the "true" values of the velocity of iodine, and some of the slight variations of velocity may be due to rain deposition effect.

3.3. Transfer Coefficient of Iodine-131 to Milk. Transfer coefficient of ^{131}I to milk (in d kg^{-1}) was calculated using the mean activity in milk (see Table 2) and in grass at the sites (Cadarache, Tricastin, St-Alban, and Agen) and also taking into account the daily ingestion of grass (3 kg d^{-1} for goats and 50 kg d^{-1} for cows). Thus we used the arithmetic mean of the daily activity in the milk divided by the daily ingested activity. One usual method employed to derive experimental transfer factors is to consider situations with the recurring administration of radionuclides to animals either in the feed or directly; the activity recovered in the milk is divided by the estimated plateau concentration of the fed and by the daily intake. In comparison with experimental assessment the methodology we used supposed that the equilibrium of ^{131}I between incorporation and excretion was reached. Concentrations in milk at Cadarache and Agen soon fell below detection limits after 20 April and after 3 May at Tricastin. The calculated arithmetic means of these coefficients are given in Table 3. This table provides also the most recent published review by Howard et al.,[15] mainly based on the critical analysis of the previous database.[16,17] Thus coefficients of ^{131}I deduced from goat's milk taken in the field strengthen the values used up to now for calculating models. The transfer coefficient to cow's milk deduced from the present study fit well with data summarized in Table 2 and with post Chernobyl data yielding a mean value of 3.4×10^{-3} d L^{-1}.[18]

Simulation models provide useful tools for quantifying this path, but the required input parameters have hardly been derived from truly in situ measurements, since the early data of Chamberlain.[19] The dry apparent deposition velocities of iodine on grass as well as milk transfer factors were deduced from the environmental monitoring carried out by the IRSN during the fallout of radionuclides related to the Fukushima's accident. Given that these values fit well with the default parameters regularly used in models, the reliability of the predicted evaluations is improved.

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