Variations in the transfer of radiocesium (^{137}Cs) and radiostrontium (^{90}Sr) from milk to cheese

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ABSTRACT

This study aimed to compare the transfer of 2 man-made radionuclides, radiocesium (^{137}Cs) and radiostrontium (^{90}Sr), from cow milk to whey and cheese in 3 different types of French cheese production with rennet coagulation. Most of the ^{137}Cs was present in the aqueous phase and became concentrated in the whey. For ^{137}Cs transfer to whey, the processing factor (Pf; i.e., the ratio of the activity concentrations) ranged between 0.86 and 1.30 ($n = 12$). The food processing retention factor (Fr), calculated using the processing efficiency, ranged between 0.85 and 1.19 ($n = 9$). No statistical difference of Pf and Fr to whey is identified for ^{137}Cs and the cheese products. The Pf calculated for ^{90}Sr transfer to cheese ranged between 3.95 and 12.16, with significant differences depending on the type of cheese. In addition, a linear correlation is observed between ^{90}Sr Pf to cheese and the Ca level in the cheese ($r^2 = 0.57$). Thus, the Pf is enhanced in hard cheeses that are enriched in calcium. This is confirmed by nearly constant Fr values, ranging between 0.66 and 0.83.

Key words: ^{137}Cs , ^{90}Sr , cheese, milk

INTRODUCTION

Radiocesium (^{137}Cs) and radiostrontium (^{90}Sr), 2 manmade radioactive nuclides with half-lives of 30.2 and 28.8 yr, respectively, have been accidentally released into the environment as a result of various events, the most recent being the contamination caused by the meltdown and explosion of the Chernobyl nuclear reactor in 1986. Before this accident, the Northern Hemisphere (more specifically, the area between latitudes 40° and 50°N) and the continent of Europe had been contaminated by radioactive fallout from nuclear testing (UNSCEAR, 2000; Renaud and Louvat, 2004; Smith and Beresford, 2005).

In meadows, the distribution of radioactive contaminants is variable depending on climatic conditions, soil types, and plant species (Albers et al., 2000; Gastberger et al., 2000; Lettner et al., 2000; Pourcelot et al., 2003; Lettner et al., 2007). McGee et al. (1995) emphasized that the greatest variation in fallout is observed on a local scale. Therefore, it is of interest to measure the radioactivity in milk and dairy products at the meadow level because this incorporates contamination on a larger scale. Milk and cheese have proved to be good indicators for evaluating radionuclide contamination over large, heterogeneous areas (Pourcelot et al., 2007). A good example of this is the correlation established between altitude and ^{90}Sr activity for one type of cheese (Froidevaux et al., 2004). However, this observation can be made because the study is specific to a single type of cheese, in this case, Emmental, which is a matured cheese.

The concentration of radionuclides in food may be affected by the food processing actions of radionuclide extraction such as boiling, the removal of certain parts of the raw food, drying, or dilution. Neglecting radionuclide losses during food processing may lead to over- or underestimation of the calculated radiation dose. Because of the variety of processes employed and products generated, this consideration has to be taken into account carefully for dairy products. It would be useful to have a better understanding and a quantitative assessment of radionuclide transfer in different types of cheese over large areas in case of accidental radioactive discharges. In particular, this would help producers select methods that minimize the transfer of contaminants to dairy products and thus to the public. It is also important to consider the transfer of these manmade radionuclides to whey, because although whey was formerly used only for animal feed, it is now used in various ways within the human food chain. Studies performed on radionuclide transfer report differences that depend on the radionuclide involved: ^{137}Cs tends to concentrate in the aqueous phase and in whey (Wilson et al., 1988; Vosniakos et al., 1989; Macasek and Gerhart, 1994), whereas ^{90}Sr tends to concentrate mainly in cheese, fol-

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lowing Ca, which is its chemical analog (Geering et al., 1990; Macasek and Gerhart, 1994). Variations in radionuclide transfer according to the type of cheese are not well evaluated. For radionuclide transfer to dairy products, IAEA (1994) distinguishes cheese (fresh or matured) and coagulation (rennet or acid) types. However, the same category shows a large range of values. For example, processing retention factor, an indicator of radionuclide transfer, ranges between 0.025 and 0.80 for ^{90}Sr in cheese matured by rennet coagulation. In addition, ECOSYS-87 (a time-dependent radioecological simulation model), modeling radionuclide transfer, does not differentiate between the various types of production to predict radionuclide transfer (Müller and Pröhl, 1993). More field values, especially for matured cheese and ^{90}Sr , would improve our understanding of radionuclide transfer to dairy products.

The aim of the present study was to evaluate the sensitivity of several cheese products to manmade radionuclides, namely ^{137}Cs and ^{90}Sr . Special attention was paid to cheese that enhanced the transfer of radioactive pollutants.

MATERIALS AND METHODS

Study Areas and the Associated Regional Cheeses

Three types of cheeses were selected from 3 regions in France. Samples of Comté and St-Nectaire were taken from the Jura and Puy-de-Dôme mountains, respectively, and samples of Coulommiers were taken from the Charente region. In each region, farms were selected for milk sampling following an altitude gradient, an indicator of the bioclimatic zone (Figure 1).

In Charente, the milk catchment area is vast, covering approximately 82,500 km² for a single cheesemaking factory. The Coulommiers produced in this region is a soft cheese with a bloomy rind formed by spontaneous syneresis, and it coagulates quickly. Acid and rennet coagulations are similar for this cheesemaking process. Despite milk being collected over large areas, 3 sites were chosen in this region because the altitude varies by only 100 to 200 m across the region. Moreover, the expected variation in manmade radionuclide activity in this western part of France is low (Roussel-Debet et al., 2007).

The St-Nectaire production area is approximately 2,000 km². St-Nectaire is a semi-hard cheese made by accelerated drainage. The principal coagulation method used is addition of rennet. The cheese is made daily on dairy farms. In this region, 3 sites were chosen at altitudes of 800, 1,000, and 1,200 m.

The Comté production area is approximately 12,000 km². Comté is a hard cheese made by accelerated

drainage. As in St-Nectaire cheese, rennet coagulation prevails. The cheese is made from milk produced in a circular area 25 km in diameter, and it ripens for more than 4 mo. The Jura mountains are composed of a series of plateaus at different altitudes. Given the large ecological differences, more attention was paid to sampling in this region, with a total of 9 sites selected from different altitudes: the plain (200 m), the first plateau (400 m), and the second plateau (800 m).

Field Sampling, Preparation, and Analysis of the Radionuclides

Eight liters (approximately 8 kg) of milk and 5 kg of cheese (total weight; **TW**) were taken from each region. Eight liters of whey were also collected from each region in the Jura and the Puy-de-Dôme. Samples were taken in each of 3 following seasons: winter (December 2006–February 2007), spring (April–June 2007), and autumn (October 2007). The minimum ripening time of 4 mo was allowed for the Comté. The cheese samples were collected in cooperation with the cheese industry so that the samples would be traceable. In the Puy-de-Dôme, cheeses are made directly after milking, so the milk and whey were collected at the same time as the cheese. In Charente, the Coulommiers and milk samples were taken at the same time from the factory and the farms, respectively. Additional information, provided since 2005 by the Institute for Radiological Protection and Nuclear Safety, Observatoire PERmanent de la Radioactivité (IRSN/OPERA) project concerning the seasonal activity of ^{137}Cs and ^{90}Sr in milk, whey, and cheese, was also used for the 1,000 m site in Puy-de-Dôme.

All samples were stored at -20°C before processing. The samples were dried at 80°C and then reduced to ash at 480°C for 52 h (Villers-Cotterets, France). The ash was then ground and compacted in a 60-mL cylindrical container before gamma-ray analysis. Gamma spectrometry was performed using coaxial hyperpure germanium detectors with a relative efficiency of more than 50%. The gamma measurements were usually taken using a counting time of 80,000 s to measure the ^{137}Cs and potassium-40 (^{40}K) activity based on peaks at 661.7 and 1,460.8 keV, respectively. For low-activity samples, counting times were increased to 160,000 and 240,000 s (Bouisset and Calmet, 1997). To measure ^{90}Sr activity, a chemical preparation of the sample was carried out and involved several stages (mineralization of 5 g of ash with 15.5 M $\text{HNO}_3\text{-H}_2\text{O}_2$ and removal of interfering elements such as Ca and Fe). Then, the strontium was separated on Sr-spec resin (crown ether chromatography resin; Eichrom Technologies, Lisle, IL). The concentration of daughter isotope, yttrium-90

Figure 1. Location of sampling sites (□) in Charente, Puy-de-Dôme, and Jura regions.

(^{90}Y), was measured using a β -proportional counter (IN20 Eurisys, St-Quentin Yvelines, France) over a period of 36 times 2 h. To compare ^{137}Cs and ^{90}Sr , activity was reported as of January 1, 2008. Calcium levels were measured by atomic absorption. All analyses were carried out in the IRSN's Environmental Radioactivity Measurement Laboratory (Orsay, France).

Statistical Analysis and Evaluation of Radionuclide Transfer

Statistical validation of the differences between activity and measured concentration was carried out using 3 types of nonparametric tests: the Wilcoxon signed-rank test for paired samples, the Mann-Whitney test for

unpaired samples, and the Kruskal-Wallis test for 3 or more unpaired samples.

Transfers of ^{137}Cs and ^{90}Sr to cheese and whey were evaluated by using 2 types of food processing transfer parameters recommended by the International Commission on Radiation Units and Measurements (ICRU, 2001). The processing factor (**Pf**) for foodstuff is the ratio of the radionuclide activity concentrations and is expressed in equation [1]:

$$\text{Pf} = \frac{[\text{Activity}_{\text{cheese or whey}}]}{[\text{Activity}_{\text{milk}}]}, \quad [1]$$

with the activities expressed as becquerels per kilogram of fresh weight (**FW**) for milk and whey samples and TW for cheese samples.

The food processing retention factor (**Fr**) is the fraction of activity of radionuclides retained in food after processing, namely the ratio of the FW of processed food divided by the weight of original raw material. Processing retention factor is expressed by equation [2]:

$$Fr = \frac{[\text{Activity}_{\text{cheese or whey}}]}{Pe \times [\text{Activity}_{\text{milk}}]}, \quad [2]$$

with the activities expressed as becquerels per kilogram of FW or TW.

An evaluation of processing efficiency (**Pe**) was made in the present study by using Ca concentrations. Cheese transfer is expressed by equation [3], and whey transfer by equation [4], with Ca concentration expressed in grams per kilogram of FW or TW:

$$Pe_{\text{cheese}} = \frac{[Ca_{\text{whey}}] - [Ca_{\text{cheese}}]}{[Ca_{\text{whey}}] - [Ca_{\text{milk}}]}, \quad [3]$$

$$Pe_{\text{whey}} = \frac{[Ca_{\text{cheese}}] - [Ca_{\text{whey}}]}{[Ca_{\text{cheese}}] - [Ca_{\text{milk}}]}. \quad [4]$$

RESULTS AND DISCUSSION

Radionuclide and Mineral Concentrations in Milk, Whey, and Cheese

Activity concentrations of ^{137}Cs , ^{90}Sr , and ^{40}K and concentration of Ca in milk, whey, and cheese samples are given in Table 1. A variation in ^{137}Cs activity level was observed in milk, ranging from 0.01 to 0.72 Bq/kg of FW. The highest activity was measured in the Puy-de-Dôme (mean = 0.19 ± 0.02 Bq/kg of FW) and the lowest in Charente (mean = 0.02 ± 0.005 Bq/kg of FW). Significant differences between the regions were demonstrated (Mann–Whitney test; $P < 0.01$). In a given region, the activity measured in milk is affected by many different factors. Activity levels are related to the intensity of radioactive pollutant deposits, which are usually increased by precipitation and are altitude-related (Renaud et al., 2003). Soil types, farming practices, and cattle diet can also increase or decrease ^{137}Cs transfer to milk (IAEA, 1994; Smith and Beresford, 2005). A small part of the contamination could be explained by the fact that the feed was not produced locally but was brought in from another region. In Puy-de-Dôme, the combination of a damp climate increasing radioactive deposits (mean annual precipitation = ap-

proximately 2,000 mm) and specific soil properties from the volcanic bedrock are likely to increase the activity in milk (Sigurgeirsson et al., 2005). Activity of ^{137}Cs in whey is of the same order of magnitude as in milk, ranging from 0.01 to 0.62 Bq/kg of FW in Puy-de-Dôme and Jura (Wilcoxon signed-rank test; $P > 0.05$; $n = 12$). The highest activity levels in whey were found in Puy-de-Dôme in farms above 1,000 m, and it was the same as for milk. In cheese, ^{137}Cs activity ranged from 0.01 to 0.52 Bq/kg of TW. Similarly, the highest activity was found in Puy-de-Dôme: on average, activity was 7 times higher than in Jura or Charente (mean = 0.15 ± 0.02 Bq/kg of TW in Puy-de-Dôme and 0.02 ± 0.01 Bq/kg of TW in Jura). ^{137}Cs activity in cheese was statistically lower than in milk (Wilcoxon signed-rank test; $P < 0.01$; $n = 28$), showing that the level of ^{137}Cs was reduced by the cheesemaking process.

The ^{90}Sr activity in milk samples from the study regions ranged from 0.01 to 0.16 Bq/kg of FW. As in ^{137}Cs , significant differences in ^{90}Sr activity were found between study regions (Kruskal–Wallis test; $P < 0.01$). The highest activity levels were at altitudes above 1,000 m in Puy-de-Dôme, where the mean activity level was 0.07 ± 0.01 Bq/kg of FW. The lowest activity levels were in Charente (altitude < 220 m), where the mean activity level was 0.03 ± 0.01 Bq/kg of FW. As for ^{137}Cs , the variability of ^{90}Sr activity in milk was the result of many factors. Aside from the initial deposits, the particular soil properties in Puy-de-Dôme may promote the transfer of ^{90}Sr to meadow vegetation, which seemed to be the case for ^{137}Cs , whereas the clay–chalk soil of the Jura may reduce the transfer. In whey, ^{90}Sr activity ranged from 0.007 to 0.02 Bq/kg of FW in Puy-de-Dôme and Jura, and there was no significant difference between the regions (overall mean = 0.01 ± 0.005 Bq/kg of FW). Activity was statistically lower than in milk (Wilcoxon signed-rank test; $P < 0.01$; $n = 13$), showing that the ^{90}Sr level in whey was reduced by the cheesemaking process. In cheese, ^{90}Sr activity ranged from 0.16 to 0.93 Bq/kg of TW. The lowest activity was measured in Charente (ranging from 0.18 to 0.24 Bq/kg of TW) and the highest at altitudes above 1,000 m in Puy-de-Dôme (ranging from 0.21 to 0.93 Bq/kg of TW; mean = 0.41 ± 0.07 Bq/kg of TW). The mean values were similar in Puy-de-Dôme and Jura, and the statistically significant difference observed for milk was not found for cheese from these regions (Mann–Whitney test; $P > 0.05$).

Potassium-40 and Ca are considered to be chemical analogs for ^{137}Cs and ^{90}Sr , respectively. In milk, ^{40}K activity ranged from 41.8 to 67.6 Bq/kg of FW with a mean of 48.8 ± 4.8 Bq/kg of FW. This variability means that no regional difference in activity can be demonstrated (Kruskal–Wallis test; $P > 0.05$). Activity

Table 1. Activities of radiocesium (^{137}Cs), radiostrontium (^{90}Sr), and potassium-40 (^{40}K) and Ca concentration in samples of milk, whey, and cheese from 3 regions and cheese productions¹

Item ²	Charente			Puy-de-Dôme			Jura		
	n	Range	Mean	n	Range	Mean	n	Range	Mean
^{137}Cs									
Milk (Bq/kg of FW)	5	0.01–0.03	0.02 ± 0.005	17	0.02–0.72	0.19 ± 0.02	20	0.01–0.08	0.03 ± 0.01
Whey (Bq/kg of FW)				9	0.01–0.62	0.23 ± 0.02	3	0.02–0.09	0.05 ± 0.01
Cheese (Bq/kg of TW)	1		0.01 ± 0.005	15	0.02–0.52	0.15 ± 0.02	11	0.01–0.05	0.02 ± 0.01
^{90}Sr									
Milk (Bq/kg of FW)	5	0.01–0.05	0.03 ± 0.01	17	0.04–0.16	0.06 ± 0.01	9	0.02–0.07	0.04 ± 0.01
Whey (Bq/kg of FW)				10	0.008–0.017	0.012 ± 0.005	3	0.007–0.014	0.01 ± 0.006
Cheese (Bq/kg of TW)	3	0.18–0.24	0.20 ± 0.05	17	0.18–0.93	0.39 ± 0.07	12	0.16–0.69	0.34 ± 0.07
^{40}K									
Milk (Bq/kg of FW)	5	47.2–55.8	50.5 ± 5.0	17	43.8–67.6	49.9 ± 5.1	21	41.8–51.7	47.6 ± 4.6
Whey (Bq/kg of FW)				10	45.4–59.9	49.2 ± 5.2	3	48.0–50.3	49.1 ± 4.4
Cheese (Bq/kg of TW)	3	34.8–36.0	35.4 ± 3.3	17	28.4–50.1	36.2 ± 3.7	12	22.6–31.2	28.4 ± 2.8
Ca									
Milk (g/kg of FW)	5	0.87–1.02	0.96 ± 0.10	16	0.74–1.02	0.89 ± 0.09	9	0.77–0.97	0.89 ± 0.09
Whey (g/kg of FW)				9	0.20–0.34	0.28 ± 0.03	3	0.25–0.29	0.27 ± 0.03
Cheese (g/kg of TW)	3	2.9–4.17	3.73 ± 0.37	13	4.92–6.59	5.55 ± 0.55	12	6.54–9.46	7.74 ± 0.77

¹The activities were reported as of January 1, 2008. The uncertainty of the mean figures is caused by measurement error.

²FW = fresh weight; TW = total weight.

of ^{40}K in whey was of the same order of magnitude as in milk, with a mean of 49.20 ± 5.0 Bq/kg of FW, whereas it was lower in cheese, with a mean of 33.2 ± 3.3 Bq/kg of TW. Significant variations in ^{40}K activity were observed between the cheeses studied (Mann–Whitney test; $P < 0.01$). Most of the ^{40}K was concentrated in the aqueous phase, thus in the whey. The lowest ^{40}K activity was measured in hard cheeses, such as Comté during pressure drainage, ranging from 22.6 to 31.2 Bq/kg of TW. The highest activity was found in cheeses such as Coulommiers during spontaneous syneresis, ranging from 34.8 to 36.0 Bq/kg of TW. Although differences were observed between the selected cheese products, the wide variation in ^{40}K for the same type of cheese shows that the whey drainage stage produces discrepancies.

Calcium levels vary little in milk. There was no statistically significant difference between regions, and mean concentrations were around 0.91 ± 0.07 g/kg of FW (Kruskal–Wallis test; $P > 0.05$). Calcium levels in whey were lower than in milk, with a mean of 0.27 ± 0.03 g/kg of FW. In cheese, our data showed variation in calcium levels, with values ranging from 2.90 to 9.46 g/kg of TW. A distinct range of calcium levels was seen for each type of cheese studied (Kruskal–Wallis test; $P < 0.001$): from 2.90 to 4.17 g/kg of TW for Coulommiers, 4.92 to 6.59 g/kg of TW for St-Nectaire, and 6.54 to 9.46 g/kg of TW for Comté. Direct relationships have been established between type of cheese and calcium content (Eck and Gillis, 2000). This is because calcium plays a decisive role in the acidification and rennet coagulation stage, forming the coagulum or curds and, thus, the cheese. During this phase, an irreversible reaction occurs, forming calcium bridges between the casein

macro-peptides. Thus, the calcium content of cheese is an indicator of the acidification and the coagulation stage and provides a way of distinguishing between different types of cheese and also of detecting variations occurring within the same process. As Geering et al. (1990) reported, most ^{90}Sr transfers to cheese through casein precipitation. Activity in cheese was statistically higher than in milk (Wilcoxon signed-rank test; $P < 0.001$; $n = 25$).

Radionuclide Processing Factors to Cheese and Whey

Radiocesium and radiostrontium Pf and Fr to cheese and whey are expressed in Tables 2 and 3. Processing efficiency, used for calculated Fr, is calculated using equations [3] and [4].

The Pf of ^{137}Cs to cheese ranged between 0.25 and 1.79. No significant difference was observed between St-Nectaire and Comté production processes (Mann–Whitney test; $P > 0.05$). The lowest and highest ^{137}Cs Pf was calculated for Comté products (Table 2).

The Pe varied among cheese types. Assuming that the Ca concentration in whey was 0.27 g/kg of FW, $\text{Pe}_{\text{cheese}}$ was, on average, 5.00 ± 1.42 for Coulommiers, 8.64 ± 2.56 for St-Nectaire, and 13.55 ± 4.04 for Comté. Thus, the amount of milk, calculated with $\text{Pe}_{\text{cheese}}$, was different according to the produced cheese (Kruskal–Wallis test; $P < 0.001$).

The Fr of ^{137}Cs to cheese ranged between 0.02 and 0.20. No difference in Fr values was found between the Comté and St-Nectaire products (Mann–Whitney test; $P > 0.05$). Coulommiers cheese showed the highest Fr

Table 2. Radiocesium (^{137}Cs) and radiostrontium (^{90}Sr) processing factor (Pf) and processing retention factor (Fr) to 3 types of cheeses

Item	Coulommiers			St-Nectaire			Comté		
	n	Range	Mean	n	Range	Mean	n	Range	Mean
$\text{Pe}_{\text{cheese}}^1$	5	3.53–6.45	5.00 ± 1.42	13	7.29–11.70	8.64 ± 2.56	6	11.37–18.69	13.55 ± 4.04
^{137}Cs Pf	2	0.39–0.69	0.55 ± 0.27	15	0.61–1.07	0.82 ± 0.19	10	0.25–1.79	1.07 ± 0.51
^{90}Sr Pf	2	3.95–4.54	4.25 ± 1.61	17	4.30–8.03	6.14 ± 1.74	6	6.43–12.16	9.32 ± 3.53
^{137}Cs Fr	2	0.10–0.20	0.15 ± 0.07	12	0.06–0.15	0.10 ± 0.03	4	0.02–0.15	0.09 ± 0.05
^{90}Sr Fr	2	0.78–0.83	0.81 ± 0.17	13	0.66–0.77	0.73 ± 0.16	6	0.66–0.73	0.71 ± 0.16

¹Processing efficiency (Pe) is evaluated from calcium concentrations in cheese and milk.

(0.10–0.20). The IAEA (1994) transfer values exhibit a similar range of variation, between 0.05 and 0.23.

Data provided no evidence of any significant correlation between Fr and Ca concentration or ^{40}K activity in cheese, although the latter was assumed to be a radiocesium analog. According to this observation, there was no enhancement of ^{137}Cs Pf in cheese with a Pf smaller than unity. Furthermore, large values of Pf and Fr recorded by a single cheese (Comté) showed that cheesemaking technology, especially manmade stages, would induce large variation of ^{137}Cs .

The Pf for ^{137}Cs to whey ranged between 0.86 and 1.30. No difference of Pf was observed between St-Nectaire and Comté production processes (Mann–Whitney test; $P > 0.05$). Regardless of the cheese produced, the Pe to whey ranged between 1.06 and 1.16. Such a short range suggests that Pe_{whey} value was globally stable and not dependent on cheese type. The Fr of ^{137}Cs to whey ranged between 0.85 and 1.19 with no statistical difference between the St-Nectaire and Comté products (Mann–Whitney test; $P > 0.05$). These values were of the same order of magnitude as those found by IAEA (1994), which ranged between 0.73 and 0.96, showing that the activity in whey is close to that in milk. The ^{137}Cs tends to concentrate mainly in the phase with the highest water content, which, in cheesemaking, is the whey. This was previously observed by Wilson et al. (1988) for a hard cheese of the Cheddar type.

The Pf range of ^{90}Sr to cheese was large, from 3.95 to 12.16. The lowest Pf to cheese was for Coulommiers,

ranging between 3.95 and 4.54. Higher values were observed for the St-Nectaire process (4.30–8.03) and higher still for the Comté process (6.43–12.16). The differences between Comté and St-Nectaire production processes are statistically significant (Mann–Whitney test; $P < 0.01$). Therefore, it seems that different cheesemaking methods affect the ^{90}Sr processing factor. The coagulation stage seems to be the key factor in the variability observed. A large Pf variation was found for the same cheese product (a factor of 2 for Comté and St-Nectaire). Seasonal variation of the microbiological and biochemical composition of milk is likely to alter the ^{90}Sr transfer for a given cheesemaking method (Eck and Gillis, 2000).

The cheese Fr for ^{90}Sr ranged between 0.66 and 0.83, with no statistical differences between the selected cheese processes (Kruskal–Wallis test; $P > 0.05$). Thus, small variations of retention factors were observed when the amount of milk needed, estimated through Ca concentration, was taken into account. Obtained Fr values fall in the high range established by IAEA (1994), which reported transfer in the cheese ranging between 0.025 and 0.80.

Thus, mass transfer regulates the ^{90}Sr concentration in cheese. As mentioned before, Ca concentration in milk was constant in the studied regions. Thus, the differences in Ca concentrations in cheese are direct indicators of $\text{Pe}_{\text{cheese}}$. A linear correlation is observed between Ca concentration in cheese and ^{90}Sr Pf (Figure 2):

Table 3. Radiocesium (^{137}Cs) and radiostrontium (^{90}Sr) processing factor (Pf) and processing retention factor (Fr) to 2 types of whey

Item	St-Nectaire whey			Comté whey		
	n	Range	Mean	n	Range	Mean
$\text{Pe}_{\text{whey}}^1$	13	1.09–1.16	1.13 ± 0.21	6	1.06–1.10	1.08 ± 0.18
^{137}Cs Pf	9	0.86–1.12	1.02 ± 0.16	3	0.95–1.30	1.11 ± 0.33
^{90}Sr Pf	10	0.16–0.27	0.21 ± 0.10	3	0.20–0.21	0.21 ± 0.16
^{137}Cs Fr	6	0.85–0.99	1.02 ± 0.28	3	0.86–1.19	1.02 ± 0.28
^{90}Sr Fr	6	0.15–0.24	0.20 ± 0.09	3	0.19–0.19	0.19 ± 0.14

¹Processing efficiency (Pe) is evaluated from calcium concentrations in cheese and milk.

Figure 2. Radiostrontium processing factor ($^{90}\text{Sr Pf}_{\text{cheese}}$) as a function of the Ca concentration in cheese. Linear regression: $y = 0.9754x + 0.927$, $r^2 = 0.53$. Cheeses: $\square$ = Coulommiers; Δ = St-Nectaire; $\circ$ = Comté.

$$^{90}\text{Sr Pf}_{\text{cheese}} = 0.975 \times [\text{Ca}]_{\text{cheese}} + 0.927,$$

where $[\text{Ca}]_{\text{cheese}}$ is given in grams per kilogram of TW, $r^2 = 0.57$.

This equation expresses the relationship between cheesemaking methods, specifically the acidification and coagulation stage correlated with the Ca level in cheese and the ^{90}Sr transfer from milk to cheese. Thus, cheeses with a high level of Ca concentration, such as hard cheese, show the highest $^{90}\text{Sr Pf}$, mainly because of the larger amount of milk needed to produce the cheese.

The Pf and Fr to whey did not strongly vary, ranging from 0.16 to 0.27 and from 0.15 and 0.25, respectively, regardless of the cheese (St-Nectaire and Comté); no statistical difference was found between the cheeses (Mann-Whitney test; $P > 0.05$). Variations of the Fr to whey, ranging between 0.20 and 0.80, were reported by IAEA (1994).

CONCLUSIONS

The measurements of ^{137}Cs and ^{90}Sr activities in milk, whey, and 3 different kinds of cheese have shown variations in the transfer of these radionuclides. No difference of ^{137}Cs transfer to cheese was observed in the cheese products. We found large Pf variations in Comté cheese, which suggest that the process used for the production of this cheese would modify ^{137}Cs transfers. The transfer of ^{137}Cs to whey was constant and independent of the type of cheese product. The fact that there was

no correlation between the $^{137}\text{Cs Pf}$ and its chemical analog (^{40}K) suggested different ways of transfer to dairy products. The $^{90}\text{Sr Pf}$ to cheese differed among types of cheeses and varied according to Ca concentration. Hard cheeses that are enriched in calcium enhance the $^{90}\text{Sr Pf}$ and thus are more sensitive than other cheeses. In the case of accidental contamination, changing the kind of cheese could be a useful way to limit radiocontaminant transfer to food.

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REFERENCES

- Albers, B. P., H. Steindl, W. Schimmack, and K. Bunzl. 2000. Soil-to-plant and plant-to-cow's milk transfer of radiocaesium in alpine pastures: Significance of seasonal variability. *Chemosphere* 41:717–723.
- Bouisset, P., and D. Calmet. 1997. Hyper pure gamma-ray spectrometry applied to low-level environmental sample measurements. Pages 73–81 in proceedings of the International Workshop on the Status of Measurement Techniques for the Identification of Nuclear Signatures.
- Eck, A., and J.-C. Gillis. 2000. Cheesemaking: From Science to Quality Assurance. Lavoisier, Paris, France.
- Froidevaux, P., J. J. Geering, L. Pillonel, J. O. Bosset, and J. F. Valley. 2004. ^{90}Sr , ^{238}U , ^{234}U , ^{137}Cs , ^{40}K and $^{239/240}\text{Pu}$ in Emmental type cheese produced in different regions of Western Europe. *J. Environ. Radioact.* 72:287–298.
- Gastberger, M., F. Steinhausler, M. H. Gerzabek, H. Lettner, and A. Hubmer. 2000. Soil-to-plant transfer of fallout caesium and strontium in Austrian lowland and Alpine pastures. *J. Environ. Radioact.* 49:217–233.
- Geering, J. J., C. Friedli, and P. Lerch. 1990. Method of determination of ^{90}Sr in the environment. *J. Trace Microprobe Techniques* 8:211–230.
- IAEA. 1994. Handbook of Parameter Values for the Prediction of Radionuclide Transfer in Temperate Environments. IAEA, Vienna, Austria.
- ICRU. 2001. ICRU report 65: Quantities, units, and terms in radioecology. *J. ICRU* 1.
- Lettner, H., P. Bossew, and A. K. Hubmer. 2000. Spatial variability of fallout caesium-137 in Austrian alpine regions. *J. Environ. Radioact.* 47:71–82.

- Lettner, H., A. Hubmer, P. Bossew, and F. Strebl. 2007. ^{137}Cs and ^{90}Sr transfer to milk in Austrian alpine agriculture. *J. Environ. Radioact.* 98:69–84.
- Macasek, F., and P. Gerhart. 1994. Distribution and speciation of caesium, strontium, and europium in the aqueous two-phase system milk-pectin. *J. Radioanal. Nucl. Chem.* 186:9–21.
- McGee, E. J., M. J. Keatinge, H. J. Synnott, and P. A. Colgan. 1995. The variability in fallout content of soils and plants and the design of optimum field sampling strategies. *Health Phys.* 68:320–327.
- Müller, H., and G. Pröhl. 1993. ECOSYS-87: A dynamic model for assessing radiological consequences of nuclear accidents. *Health Phys.* 64:232–252.
- Pourcelot, L., D. Louvat, F. Gauthier-Lafaye, and P. Stille. 2003. Formation of radioactivity enriched soils in mountain areas. *J. Environ. Radioact.* 68:215–233.
- Pourcelot, L., P. Steinmann, and P. Froidevaux. 2007. Lower variability of radionuclide activities in upland dairy products compared to soils and vegetation: Implication for environmental survey. *Chemosphere* 66:1571–1579.
- Renaud, P., and D. Louvat. 2004. Magnitude of fission product depositions from atmospheric nuclear weapon test fallout in France. *Health Phys.* 86:353–358.
- Renaud, P., L. Pourcelot, J.-M. Métivier, and M. Morello. 2003. Mapping of ^{137}Cs deposition over eastern France 16 years after the Chernobyl accident. *Sci. Total Environ.* 309:257–264.
- Roussel-Debet, S., P. Renaud, and J. M. Métivier. 2007. ^{137}Cs in French soils: Deposition patterns and 15-year evolution. *Sci. Total Environ.* 374:388–398.
- Sigurgeirsson, M. A., O. Arnalds, S. E. Palsson, B. J. Howard, and K. Gudnason. 2005. Radiocaesium fallout behaviour in volcanic soils in Iceland. *J. Environ. Radioact.* 79:39–53.
- Smith, J. and N. A. Beresford. 2005. *Chernobyl: Catastrophe and Consequences*. Springer, New York, NY.
- UNSCEAR. 2000. Report to the General Assembly, Sources and Effects of Ionizing Radiation. United Nations Scientific Committee on the Effects of Atomic Radiation, New York, NY.
- Vosniakos, F., P. Karakoltsidis, A. Mountzis, A. Kesidou, S. Ganatsios, and A. Bizopoulos. 1989. Transfer of ^{121}I and ^{137}Cs from cow milk to cheese and other products. *Aust. J. Dairy Technol.* 44:44–46.
- Wilson, L. G., R. C. Bottomley, P. M. Sutton, and C. H. Sisk. 1988. Transfer of radioactive contamination from milk to commercial dairy products. *J. Soc. Dairy Technol.* 41:10–13.