

## Radiocesium Contamination in Samples of Blueberries Jams Collected in Stores of NE Italy (2013-2017)

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### Abstract

The monitoring of radioactivity in foodstuffs is carried out for the purposes of food safety in order to follow the evolution of the contamination as result of incidents that occurred both in the past (eg Chernobyl) and in more recent times (eg Fukushima). Then, the movement of goods from these countries to European Union may cause the propagation of foods potentially toxic for health.

At the Port of Trieste, in the period of September 2013, some loads of blueberries (*Vaccinium mirtyllus* L.) were examined within the application of EC Regulation 733/2008, on the conditions governing imports of agricultural products originating in the countries affected by the nuclear accident of Chernobyl in 1986.

The fruits came from Ukraine and were intended for the food preparations containing blueberries, in particular jams and marmalades.

As in some cases significant values of Radiocesium concentrations (Cesium-137) were found in blueberries, though below the limit of 600 Bq/kg as required by the Regulations, it was decided to verify whether the presence of this contaminant in the raw material at the tested levels could interest the finished products. In this work, we have therefore collected and analyzed some samples of concentrated blueberries products in different large stores, in order to verify the magnitude of the possible contamination by radio Cesium.

**Keywords:** Blueberries, contamination, EC Regulation 733/2008, jams, Radiocesium, gamma spectrometry

### 1. Introduction

The plant of Blueberry (*Vaccinium* sp.) belongs to the family of Ericaceae and is characterized by a root system called "rhizome". The rhizome grows horizontally within the first few inches of the organic layer of the soil even for many meters. It is formed by endotrophic-mycorrhiza that possess a high capacity for mobilization and absorption of mineral salts from the soil, similarly to many species of mushrooms. This property is common to all Ericaceae plants and can lead sometimes to high values of radio-contamination from Cs-137.

It has already been observed in the past that the fruits of Blueberry may show concentration values of radio-caesium even greater than those recorded in different species of edible mushrooms (Zappa et al. 2001). It has been shown that the Blueberries plant can also be considered as an indicator of air pollution (Deyeva & Gerasimenko, 1994) and environmental pollution in general, as well as for other metals (Deyeva & Maznaya, 1992).

Cs-137 is considered as the more persistent tracer in the environment following cases of radiocontamination such as nuclear tests in previous years (Fig.1) and different types of nuclear power plants accidents (e.g. Chernobyl and Fukushima).

After about 15 years from the environmental intake, the contribution to the residual gamma dose is due for almost 100% to Cs-137 nuclide (half-life 30 years). As concerns other radioactive isotopes, the most persistent synthetic isotope presents in the fall-out but at much lower level is Sr-90, with a half-life of about 29 years. Therefore, the environmental monitoring of the long-term consequences (over 15 years) of fall-out or nuclear accidents concerns the measurement of Cs-137 and, in some cases, also of Sr-90.

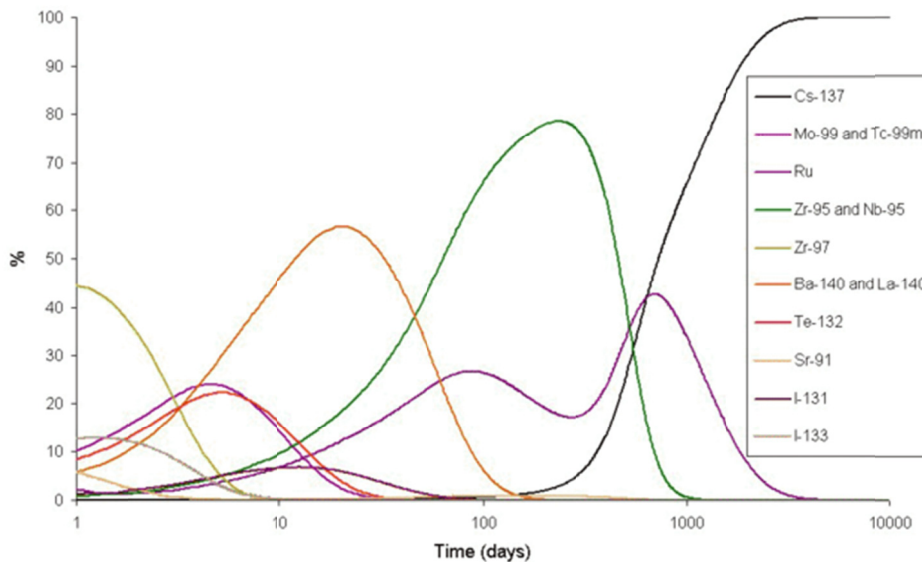


Figure 1. The relative contribution to the gamma dose rate due to the main isotopes in bomb fallout (Font: <http://en.wikipedia.org/wiki/Image:Bombfalloutisotopicsig.png> 22 July, 2006)

It is to note that the cited EC Regulation (2) reports the following limits for the subjected foods (Article 2):  
 “The accumulated maximum radioactive level in terms of caesium-134 and -137 shall be:

- a) 370 Bq/kg for milk and milk products listed in Annex II and for foodstuffs intended for the special feeding of infants during the first four to six months of life, which meet, in themselves, the nutritional requirements of this category of persons and are put up for retail sale in packages which are clearly identified and labeled ‘food preparation for infants’;
- b) 600 Bq/kg for all other products concerned.” (Reg. CE 1609/2000),

The products subjected are reported in the following Table 1

Table 1. List of products covered by Regulation (EEC) No. 737/90 (Annex to Regulation (EC) no. 1609/2000 of 24 July 2000

CE cod. n.	Products
0101 19 10	Live horses, other than pure-bred breeding, for slaughter
0102 90	Live bovine animals, other than pure-bred breeding, domesticated species
0103 91	Live swine, other than purebred breeding, weighing less than 50 kg
0103 92	Live swine, other than purebred breeding, weighing more than 50 kg
0104 10	Live sheep, other than pure-bred breeding
0104 20 90	Goats live, other than pure-bred breeding
0105	Live poultry, that is to say, fowls of the species Gallus domesticus, ducks, geese, turkeys and guinea fowls
0106 00	Other live animals
Chapter 2	Meat and edible offal
ex Chapter 4	Dairy products; birds' eggs; natural honey; edible products of animal origin, not elsewhere specified or included, except for products falling within CN codes 0408 11 20 and 0408 19 20 and 0408 99 20
ex 0709 51	Mushrooms, fresh or chilled, other than cultivated mushrooms
ex 0710 80 69	Mushrooms (uncooked or cooked by steam or water), frozen, other than cultivated mushrooms ex 0711 90 60 mushrooms provisionally preserved (for example, sulfur dry mushrooms in brine, in sulfur water or in other preservative solutions), but unsuitable in that state for immediate consumption, other than cultivated mushrooms
0712 30 00	Dried mushrooms, whole, cut, sliced, broken or in powder, but not further prepared, other than cultivated mushrooms

Table 1 (continue)

CE cod. n.	Products
0810 40	Blueberries, cranberries blacks and other fresh fruits of the genus <i>Vaccinium</i>
0811 90 50	Fruit of the species <i>Vaccinium myrtillus</i> , uncooked or cooked in water or steamed, frozen, whether or not containing added sugar or other sweetening matter
0811 90 70	Fruit of the species <i>Vaccinium angustifolium</i> and <i>Vaccinium myrtilloides</i> , raw or boiled in water or steamed, frozen, whether or not containing added sugar or other sweetening matter
0812 90 40	Fruits of the species <i>Vaccinium myrtillus</i> , preserved (for example in sulfur dioxide, water solutions of sulfur or other preservative solutions), but unsuitable for immediate consumption
1601 00	Sausages and similar products, of meat, meat offal or blood; food preparations based on these products
1602	Other prepared or preserved meat, meat offal or blood
ex 1603 00	Extracts and juices of meat
ex 2001 90 50	Mushrooms, prepared or preserved by vinegar or acetic acid, other than cultivated mushrooms
ex 2003 10 80	Mushrooms, prepared or preserved otherwise than by vinegar or acetic acid, other than cultivated mushrooms

## 2. Material Studied

Six batches of black blueberries (*Vaccinium myrtillus* L.) were measured for radiocaesium at the Port of Trieste (Northern Italy) in September 2013, during the checks required on the imported food products from non-EU countries. The batches, loaded on trucks, were coming from Ukraine and directed to a food industry of the province of Bologna in the Emilia Romagna region in the middle of the Italy.

The data obtained for the specific activity for the only Cs-137 (the isotope Cs-134 is no longer detectable with the sensitivity of the used method, as the half-life is 1.9 year) are reported in the table 2.

Table. 2 Values of specific activity of Cs-137 found in the blueberries batches in imported foodstuff at the Port of Trieste in September 2013.

n.	date	Goods	Quantity (kg-net weight)	Cs-137 (Bq/kg)
1.	10/09/2013	<i>Vaccinium myrtillus</i> L	20.000	350 ± 5%
2.	10/09/2013	<i>Vaccinium myrtillus</i> L	20.000	≤ 5.0
3.	17/09/2013	<i>Vaccinium myrtillus</i> L	20.000	49 ± 10%
4.	24/09/2013	<i>Vaccinium myrtillus</i> L	20.000	≤ 5.0
5.	25/09/2013	<i>Vaccinium myrtillus</i> L	20.000	≤ 5.0
6.	30/09/2013	<i>Vaccinium myrtillus</i> L	20.000	43 ± 15%

The samples analyzed showed that three of the six batches showed values lower or equal to the sensitivity limit of the method (<0.5 Bq/kg) while only in one of the lots was found a value closer to the maximum limit (370 Bq/kg) required by the EC Regulation 733/2008 for the dietetic products for children (370 Bq/kg).

For this reason, we decided to undertake the present study on concentrate Blueberries products, like jams and juices. The sampling, even if limited to a small number of products was performed in large stores both of the city of Trieste and Padua, and interested some of the most diffused trademarks of these products in Italy.

## 3. Methods and Technique

In this work, we have collected and analyzed different samples of concentrated blueberries products in different large stores.

The determination of the Cs-137 specific activity was performed by gamma spectrometry on the untreated sample, put in geometry in a Marinelli becker of 500 ml of volume. The measurement system consists of the following modules: coaxial detector with probe LaCl<sub>3</sub> 1,5'x 1,5' 38B38 n.s. SFC-334R Scionix (*Scionix Holland B.V - Bunnik, Netherlands*) and multichannel analyzer. PX4- Amptek (*Amptek, Inc. – Bedford (MA) – USA*). The spectrometric chain is equipped with the analytical software "Amptek - Pmca" for the quantitative analysis of Cs-137 at 662 keV and was inter-calibrated with the high-resolution gamma spectrometry with high purity germanium detector systems of the laboratory ICMATE-CNR in Padua. To ensure an adequate sensitivity at the measurement, the acquisition time was set at about 42000 seconds (12 hours) for each sample. In these conditions the sensitivity limit of the method was calculated in 5.0 Bq/kg for the Cs-137.

#### 4. Results and Discussion

The values of the Cs-137 concentration (or specific activity), with the relative standard deviation in percentage, found in the analyzed samples are reported in the following table (Table 3), in the above cited conditions. Each value is the average of four different determinations on the same sample.

Table. 3 Values of specific activity of Cs-137 found in blueberries preparations and estimated specific activity in the original fruits.

Product (trademark as “,”)	Place of production	Cs-137 (Bq/kg product)	Cs-137 (Bq/kg fruit)
Organic Juice “Bio - Nettare di Mirtillo – Biologico”	Austria	≤ 5.0	nd
Dried Cranberries “Mcennedy – American Way”	Germany	≤ 5.0	nd
Organic Jam “Fiordifrutta – biologico Rigoni di Asiago”	Italy - Asiago (VI)	≤ 5.0	nd
Jam Gilli	Italy – Cardano (BZ)	≤ 5.0	nd
Organic Jam “Scelta Verde Despar – Biologico”	Italy – Casalecchio di Reno (BO)	184 ± 8%	307
Jam “Hero – Light”	Germany	47 ± 27%	94
Jam “Maribel”	Germany	≤ 5.0	nd
Jam “Natureta – Potocco”	Slovenia	175 ± 8%	389
Jam “Natturrein – d’Arbo”	Italy – Bozen	88 ± 13%	176
Jam “Sarchio”	Italy – Carpi (MO)	≤ 5.0	nd
Jam “St. Dalfour”	France	≤ 5.0	nd
Jam “Vis – Natura Golosa”	Italy – Lovero (SO)	≤ 5.0	nd
Organic Jam “Vivi Verde – biologico Coop”	Italy – Codigoro (FE)	112 ± 11 %	187

The EU regulations use the term “cumulated radioactivity”, meaning by this the contribution of the Cs-134 isotope to the total radioactive Cesium. This isotope was present in a ratio 1:2 at the time of the Chernobyl accident (1986), but given its rapid half live (less than two years) it is no more detectable in food given the sensitivity of our method. To detect Cs-134 nowadays it would be necessary to use high resolution spectrometric systems, but its determination appears to be meaningless both from the point of view of the safety and the quality of food products.

The data reported in table 4 as “Cs-137 (Bq/kg fruit)” have been obtained by the ratio of the measured value with the percentage of fruit declared on the label. As it can be observed many of the data show low concentration values of Cs-137, always under the limit value of 600 Bq/kg. In eight out of thirteen cases the values are under the sensitivity limit of the method. The higher results (over 100 Bq/kg) have been recorded in one case in a jam produced in Slovenia and in other two cases in zone of the Emilia Romagna region. This fact could be fortuity or not, but the batches of blueberries detected at the Trieste Port in September 2013 were just directed to this region in the Central Italy.

Even more recent data of samples collected in the period 2015-17 seem to confirm this supposition (Table 4).

Table. 4 Values of specific activity of Cs-137 found in blueberries preparations and estimated specific activity in the original fruits (period 2015-2017)

Product (trademark as “,”)	Place of production	Cs-137 (Bq/kg product)	Cs-137 (Bq/kg fruit)
Jam “Pam Panorama”	Spinea (VE) - Italy	81±10%	147
Jam “Alce Nero”	Monterenzio (BO) -Italy	122±10%	120
Jam “Rigoni di Asiago”	Forza (VI) - Italy	66±10%	120
Jam “Maribel”	Weinfelden – Suisse	<3	< 3
Jam Bio “Sarchio”	Carpi (MO) - Italy	124±10%	225
Organic Jam “Mirtilli biologico”	Cesena (FC) - Italy	148±10%	135
Jam “Conad – Bio”	Novaledo (TN) - Italy	22±10%	44
Jam “Zuegg”	Werneuchen - Germany	21.5±10%	43

## 5. Conclusion

The results of this work, even if limited to a small number of samples, show that in this food products Cs-137 amounts are still present in some cases at well detectable levels. For these reasons we believe it is important to maintain the control system on the interested foodstuffs (Table 1) currently prolonged by the EU Council until 2020 (*Council Regulation EC 1408/2009*). Even if there is no evidence of any sanitary risk, the interest of this work may concern especially the food quality. The detection of radio cesium in products defined as “organic” could suggest the use in their production, as raw material, of fruits imported from countries of East Europe, rather than the use of local controlled fruits.

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