Research Article

Study of radiocesium contamination from Chernobyl accident in samples of black blueberry jams

Chiara Cantaluppi1*, Daniele Zannoni2 and Massimo Calabrese3

1CNR-ICMATE, University of Padua, 35127, Italy
2CNR-ISP, Via Torino 155, Venice, 30172, Italy
3DEAMS, Merchandise Laboratory, University of Trieste, Via A. Valerio, 6, I-34127 Trieste, Italy

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*Corresponding author: Chiara Cantaluppi, Institute of Chemistry of Advanced Materials and Technologies for Energy, National Research Council, Research Area of Padua, Padua, Italy. Tel: +39498299968, E-mail: chiara.cantaluppi@cnr.it

ORCID: https://orcid.org/0000-0002-0476-189X

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Abstract

More than 30 years after the Chernobyl accident, foodstuff based on black blueberries (Vaccinium myrtillus) with relevant 137Cs activity concentrations are still found in the European Union market. Recently, mass media reported that food products based on black blueberries produced in UE were rejected by Asian markets because the 137Cs activity concentration was greater than 100 Bq kg⁻¹. It is known that Ukraine, Belarus and Russia are great exporters of black blueberries: there is a strong suspicion that the berries are collected also from the most radio-contaminated areas of these countries and introduced in UE markets.

For this reason, about 40 samples of black blueberry preserves as jams, marmalades, stewed fruits were analysed with high resolution gamma spectrometry to measure 134Cs and 137Cs activity concentration. All the food preparations of the study have been collected in supermarkets and in local stores of northern Italy. 134Cs was less than minimum detectable activity in all samples, as expected following its half-life. On the other hand, 137Cs activity concentration was widely variable among samples: the minimum, mean and maximum concentration were 1, 54 and 162 Bq kg⁻¹, respectively. The activity concentrations found in these products were corrected for the radioactive decay and reported to the same date. The activity concentration in fruits used for preserves was calculated from fruit percentage content reported on the product label. In fruits used for marmalades, jams and stewed fruits, 137Cs activity concentration up to 230 Bq kg⁻¹ was found. In a sample of blueberries in syrup, 137Cs activity concentration in blueberries was 450 Bq kg⁻¹.

From statistical analysis and contamination data of European soil, it is clearly seen that the contamination from Chernobyl accident is still relevant in some forest ecosystems and its transfer to some vegetables must be taken in account. Organic and non-organic jams show no significant difference in terms of mean 137Cs concentration. However, a potential multimodal distribution is observed for organic products, highlighting the possibility of fruit harvesting in areas with different 137Cs soil concentration. The results of this study are aimed to expand the current knowledge on the distribution of 137Cs in black blueberry products, allowing health organizations to improve the absorbed dose budget from raw berries-based food.

Introduction

It is well known that after the Chernobyl Nuclear Power Plant (NPP) accident, large areas of many European countries were significantly contaminated by radioactive fallout. In particular, besides Ukraine and Belarus (countries closer to the accident site), also wide areas of the Russian Federation, Scandinavia, Bulgaria, Austria and some spots in the Alps were affected by the deposition of radioactive material [1-3] (Table 1, Figure 1). The most highly contaminated area around Chernobyl, with 137Cs soil contamination higher than 185 kBq m⁻², is about 30 000 km² wide: these affected territories are mainly rural (agricultural and wide forests).

In natural and semi-natural ecosystems, as forests and wooded areas, a persistent recycling of radiocesium is often observed. In fact, wooded areas hold back radionuclides from atmospheric fallout and recyle them in a continuous cyclic...
exchange between upper soil layers, bacteria, microfauna, microflora and vegetation. Moreover, some mountain areas with high rainfall mean rate (e.g. in the Alps) had high wet deposition of the Chernobyl fallout [4].

Hence, although $^{137}$Cs contamination decreased in many agricultural products, it is still currently present in spontaneous mushrooms, in berries and in wild animals’ meat because of the persistent contamination of forest ecosystems. By the way, in recent years the consumption of blueberries is continuously growing, also because berries are considered rich in nutraceutical substances.

The black blueberry plant (*Vaccinium myrtillus*) belongs to the ericaceous family and is characterized by a root system called “rhizome”. The rhizome grows up horizontally in the first, organic layer of the soil, with a few centimetres thickness and extending in distance also for some meters. It is made up of “endotrophic mycorrhiza” that have a high mobilization capability and high capability of absorption of mineral salts from the soil, similarly to many species of mushrooms. This property is common to all ericaceous plants and may sometimes give rise to high $^{137}$Cs concentration values [5].

In Italy, the black blueberry plant grows spontaneously in the Alps and in part of the Apennines, in acid and loose soils; the fruits are usually collected by hand. Moreover, in Italy blackberry cultivation is almost completely family-run, for domestic consumption: industrial cultivation of blackberry is almost absent; they are indeed present in northern Europe,
where the climate is more favourable. The blackberries used by the Italian food industries are imported; they come mainly from northern Europe and from ex URSS Federation countries. Eastern Europe countries are increasingly investing in the production and export of berries, mainly intended to European Union market. Therefore, it makes sense that berries with high radiocesium concentration could have been harvested from local rural population and from the most contaminated areas of Belarus, Ukraine and western Russia [6].

Following the accident of Chernobyl NPP, controls on agricultural goods for human consumption produced in third countries respect to European Union are regulated by Regulations and Recommendations of the European Union. As it is known, the CE regulation n. 733/2008 [7] establishes the following upper limits for the sum of the activity concentrations of $^{134}$Cs and $^{137}$Cs, at 370 Bq kg$^{-1}$ for dairy products and for baby food, and at 600 Bq kg$^{-1}$ for meat, milk, honey, spontaneous mushrooms, blackberries. However, CE Regulation N. 1048/2009 [8], that modifies the previous regulation, establishes that controls ended on 2020, March 31$^{\text{st}}$.

From 2013 to 2017 the authors analysed samples of black blueberries at customs of the Trieste harbour coming from loads imported from Ukraine [9]. In one sample of black blueberries $^{137}$Cs was found up to 350 Bq kg$^{-1}$. The high variability and sporadic high concentration of $^{137}$Cs in raw berries样品 leads to question how the accumulated $^{137}$Cs is then transferred to market-available foodstuff. For this reason, the authors from 2013 to 2019 collected and analysed jams, marmalades, stewed fruits and other foodstuffs based on blue blackberry. They were analysed with high resolution gamma spectrometry for the determination of $^{137}$Cs activity concentration. In this paper we are reporting and discussing results for foodstuff bought in supermarkets in 2017, 2018, 2019, mostly jams and stewed fruits.

Material studied

The jars of jams and other foodstuffs based on blackberry were bought in supermarkets and local stores in Veneto and Friuli Venezia Giulia regions, northeast Italy. Unfortunately, on product label the geographical origin of the fruits is not reported. All the results of $^{137}$Cs activity concentration were corrected for radioactive decay recalculated at the date of 2019, January 1$^{\text{st}}$.

Methods and technique

Sample preparations and analysis were done in the CNR-ICMATE laboratories of the Italian National Research Council. From the original container of foodstuff, about 50 ml were sampled, weighted and put in a plastic jar measurement geometry of 50 ml volume without any other sample pre-treatment. Before the extraction of enough subsample for the measurement geometry, each marmalade was thoroughly mixed to ensure sample homogeneity. Then, the plastic jar was put on the top of the entrance window of the detector and analysed through high resolution gamma spectrometry using an high purity germanium detector (HPGe) with Berillium entrance window. The detector’s characteristics are reported in Table 2. The detector is installed inside a lead well, internally coated with a copper and cadmium sheets to achieve a very low environmental background. The software Genie2000® (Canberra TNE) was used for spectral acquisition and quantitative spectral analysis. Nuclide library used is Nucleide Lara LNHB (2018).

The efficiency calibration for the measurement geometrical configuration (jar 50 ml) was calculated with multi–gamma certified standard solutions (QCY48 e QCYB40 by Amersham) in the same geometrical configuration as the measurements. The results were corrected for “matrix effect” considering the different density and composition of the samples with respect to the standard solution used for efficiency calibration. The efficiency of the measurement system is periodically checked through inter–calibration tests promoted by National Physics Laboratory (Teddington, UK). Acquisition times for the analysis of $^{137}$Cs were variable from one sample to another and were manually chosen with the aim to achieve a statistical counting uncertainty (1 Standard Deviation SD) in the $^{137}$Cs photopeak at 661,7keV of less than 5%. The Limits of Detection (LOD) were estimated at 95% confidence level.

Results

Jams, marmalades and stewed fruits

Measurements results and activity concentrations of $^{137}$Cs in blackberries are reported in Table 3. As predictable, $^{137}$Cs was not detected in any sample (always under the detection limits of 1Bq kg$^{-1}$). We can reasonably argue that $^{137}$Cs present in the samples is attributable mostly to the Chernobyl fallout. In fact $^{137}$Cs soil inventory due to weapon tests in the atmosphere is meanly about 2kBq m$^{-2}$ [1]. For what concerns Fukushima accident in Europe the contribution to total $^{137}$Cs might be not relevant [2,10].

The mean $^{137}$Cs activity concentration of 36 samples of blueberry jam bought in supermarkets and local stores was 54 ± 50 (1 SD) Bq kg$^{-1}$ and the maximum activity recorded was 162Bq kg$^{-1}$. Percentage content of fruits declared on product label for all products (organic and non–organic) was in the range 50%–60%, except two products declaring respectively

| Material studied |
|------------------|
| Jams, marmalades and stewed fruits |

| Table 2: Principal features of the detector used. |
|-----------------------------------------------|
| DETECTOR | RG – 1 |
| Producer | Canberra |
| Diameter (mm) | 53.5 |
| Thickness (mm) | 53.0 |
| Distance from detector and window (mm) | 5 |
| FWHM (@122 keV) | 0.548 |
| FWHM (@1332 keV) | 1.73 |
| Peak/Compton ratio | 55.9/1 (@1332 keV) |
| Relative efficiency (%) | 25.3% (@1332 keV) |
| Depletion voltage (V) | -2500 |
| Working voltage (V) | -4000 |

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a fruit content of 102% and 110%. The mean fruits content for organic and non-organic products is very similar (64% and 60%, respectively).

The $^{137}$Cs content in blackberries was calculated starting from the fruit content declared on the label (Table 3, column 5), resulting in a mean and maximum $^{137}$Cs activity in the fruits of $85 \pm 72$ (1 SD) and $234\text{Bq kg}^{-1}$, respectively (Table 4).

To evaluate if cultivation methods could influence $^{137}$Cs content, the whole marmalade dataset was divided in two sub-samples: one of "organic" jams (19 samples), the other

Table 3: Activity concentrations of $^{137}$Cs in product and blueberries. Measurement error reported as 1 SD.

| Brand/Producer | Organic label | Fruit's origin | Country of production | Berries content (%) | $^{137}$Cs activity concentration in product (Bq kg$^{-1}$) measured | Measurement error (Bq kg$^{-1}$) | $^{137}$Cs blackberries activity concentration* (Bq kg$^{-1}$) calculated |
|----------------|---------------|----------------|-----------------------|---------------------|-----------------------------------------------------------------|---------------------------------|-----------------------------------------------------------------|
| Orogel (Auchan) IT IT - FC | 70 | 162 | 5 | 232c |
| Solo Frutta Mirtilli Bio IT BIO 007 IT - AR | 110 | 160 | 3 | 145 |
| VIS IT BIO 009 IT - SO | 70 | 134 | 4 | 191 |
| Sarchio IT BIO 009 IT - MO | 55 | 129 | 4 | 234 |
| Alce Nero IT BIO 009 UE/Non UE IT - BO | 102 | 124 | 3 | 122 |
| Orogel (SIGMA) IT BIO 009 IT - FC | 55 | 112 | 4 | 204 |
| Agrimontana ** IT - CN | 80 | 99 | 4 | 123 |
| Pam/Panorama (1/2) IT BIO 009 IT - FC | 55 | 81 | 3 | 147 |
| Horvat Wilhelm SRL (1/2) UE IT - BZ | 60 | 79 | 3 | 131 |
| Boschetti IT BIO 009 UE/Non UE IT - VR | 60 | 78 | 3 | 130 |
| L'Ape IT BIO 002 IT - VI | 52 | 75 | 2 | 145 |
| L’ape di Cardin (1/2) IT BIO 014 UE/Non UE IT - SA | 52 | 75 | 3 | 145 |
| Despar (1/2) IT - FE | 52 | 72 | 2 | 139 |
| L’ape di Cardin (2/2) IT BIO 002 UE/Non UE IT - VI | 52 | 70 | 3 | 134 |
| Rigoni di Asiago (1/2) IT BIO 007 UE/Non UE IT - VI | 55 | 69 | 3 | 125 |
| D'Arbo A IT - CB | 70 | 60 | 4 | 86 |
| Santa Rosa IT - VR | 52 | 33 | 2 | 65 |
| GTC SRL (Sane Bontà) IT BIO 007 IT - TO | 55 | 32 | 2 | 57 |
| Orto d’Autore IT - CB | 70 | 27 | 2 | 38 |
| Rigoni di Asiago (2/2) IT BIO 007 UE/Non UE IT-Vi | 55 | 27 | 2 | 49 |
| Despar (2/2) IT BIO 006 UE/Non UE IT - TN | 55 | 25 | 0.5 | 46 |
| Conad IT - TN | 50 | 23 | 2 | 46 |
| Zuegg DE | 50 | 22 | 0.9 | 45 |
| Maxi Di IT BIO 007 UE/Non UE IT - TN | 60 | 22 | 2 | 36 |
| Luigi Lazzaris e Figlio SRL IT BIO 007 IT - TV | 65 | 15 | 1 | 24 |
| Hero Italia SPA ES | 50 | 4 | 1 | 7 |
| Primizieparis IT BIO 007 UE/Non UE IT - TO | 55 | 3 | 0.7 | 5 |
| Bonne Maman FR | 50 | 3 | 0.3 | 5 |
| Eurofood IT BIO 014 UE/Non UE IT - TO | 55 | 3 | 0.1 | 5 |
| Horvat Wilhelm SRL (2/2) UE IT - BZ | 60 | 2 | 0.5 | 3 |
| Cadoro IT BIO 007 UE/Non UE IT - TO | 55 | 2 | 0.4 | 4 |
| Gilli IT - BZ | 60 | 1 | 0.2 | 2 |
| Consilia IT - VR | 70 | 1 | 0.5 | 2 |
| Selex IT - TN | 50 | 1 | 0.7 | 2 |
| Pam/Panorama (2/2) BE | 55 | <4 | 5 | 4*** |
| Maribel (UDL) IT BIO 007 DE | 50 | <3 | 3 | 3*** |

* Blackberries activity calculated accounting for the fruit % content declared on the product label.
** Accounted for non-organic product but fruit declared as “wild berries”.
*** Estimated from LOD/2.
one of “non-organic” jams i.e. jams without any information about fruits cultivation (17 samples). The results of the descriptive statistics are reported in Table 4. Mean $^{137}$Cs activity concentration was higher in organic jams (98) respect to non-organic jams (70) but standard deviation and maximum value were almost equal.

Data distributions of the two sub-samples are reported in Figure 2. It is worth to be noted that for organic products and fruits the distribution is probably multimodal: about half samples have $^{137}$Cs activity concentrations less than 90Bq kg$^{-1}$, similarly for organic products and for non-organic products. Fruit frequency distribution for organic product shows a possible mode between 140 and 150Bq kg$^{-1}$.

The two sub-sample distributions were compared through parametric and non-parametric statistical tests. The use of different tests is for minimizing biased interpretation due to non-normal and skewed distribution and thus avoiding type II error. The two-sample $t$-test was used as the parametric test (H0: data from subsamples come from independent random samples with equal means and equal but unknown variances) while the Wilcoxon rank sum test was used as the non-parametric test (H0: data from subsample sets come from continuous distributions with equal medians) that is considered a non-parametric alternative to the two-sample $t$-test. Such tests have different power when they deal with distributions with different shape. The $t$-test, for instance, is more powerful for symmetric distributions with low sample size. On the other hand, the Wilcoxon rank sum test is more powerful with distributions that show large skewness [11]. As can be noted in Table 5, both parametric and non-parametric statistical tests suggest to not reject H0, leading to the conclusion that organic and non-organic products and fruits come from similar distributions (equal means and equal medians). However, for organic fruits the possibility of two distinct distributions seems plausible: one subsample set with activity concentration $\leq 50$Bq kg$^{-1}$ (n=8) with a mean activity concentration (±1SD) of 21±20Bq kg$^{-1}$ and the other one (n=8) with a mean activity concentration (±1SD) of 137±10Bq kg$^{-1}$. Due to the large relative number (42%) of blackberries in the cluster centred at 137Bq kg$^{-1}$ and their low variability in $^{137}$Cs content (~7%) it seems reasonable to assume that the berries of such cluster comes from the same area (or from areas with similar $^{137}$Cs content in soil). It is worth noting that both the statistical tests used here have low power when dealing with multimodal distributions. This means that the test result for $^{137}$Cs content in fruits must be considered cautiously.

**Other blackberry-based foodstuff**

Also a canned fruit sample “Frutta Sciroppata” VIS (IT-SO) bought in supermarket was analysed (not reported in Table 3). The product is made of blackberry fruits in sugar syrup. For the good as it is (fruits submerged in syrup), the activity

| Table 4: Descriptive statistics of the $^{137}$Cs content in blueberries jams, after separation of organic and non-organic products. |
|-------------------------------------------------------------|
| All jams | Organic jams | Non-organic jams |
| Number of samples | 36 | 19 | 17 |
| $^{137}$Cs Mean activity concentration (Bq kg$^{-1}$) | 85 | 98 | 70 |
| Standard deviation (Bq kg$^{-1}$) | 72 | 71 | 74 |
| Minimum value (Bq kg$^{-1}$) | <2 | <2 | <2 |
| Maximum value (Bq kg$^{-1}$) | 234 | 234 | 232 |
| Median (Bq kg$^{-1}$) | 48 | 123 | 45 |
| Curtosys | 0.7 | 0.18 | 0.95 |

**Figure 2:** Box plots for $^{137}$Cs activity concentration in Organic and Non-Organic products and fruits. Boxplot parameters: box is perc. 25,75; diamond is the mean; boundaries are min and max values. An attempt of log-normal distribution based on observed frequency classes is reported on the left of each boxplot.
concentration measured was 168±5Bq kg⁻¹. Since this is quite a high 137Cs concentration, on the reasonable hypothesis that radioesium was initially contained in the berries and not in the sugar syrup, an attempt was made to calculate the initial activity concentration in the berries. At first, the berries were manually separated from the syrup. An amount of the canned berries was then drained off and repeatedly rinsed in milliQ water, with the aim to remove possibly all the syrup from the berries. The 137Cs activity concentration for the washed berries, for the syrup and for the rinsing water were measured and results are reported in Table 6. From these data and from the weight of the analysed samples, the initial activity concentration of 137Cs in the berries (A*) was re–calculated assuming that 137Cs were initially present only in the berry using equation 1:

\[
A^* = \frac{A_i m_b + A_s m_s}{m_b}
\]

(1)

where \(A_i\) and \(m_s\) are the 137Cs activity concentration and mass of rinsed berries, respectively, and \(A_s\) and \(m_s\) are the activity and mass of the syrup analysed separately. For completeness, also the washing water was collected and analysed: the activity concentration was 5±1Bq kg⁻¹. The results are reported in Table 6. The value obtained from this trial is obviously only semi-quantitative, as it was not possible to collect and analyse all washing water and separation of syrup and berries was not 100% efficient. Anyhow the result of the activity concentration of 137Cs in the berries is quite high: 456Bq/kg: this supports the thesis that many lots of blueberries come from highly contaminated regions.

Discussion

The results of the activity concentration of 137Cs in blackberry jams found in this work are higher than those reported in recent literature [12,13]. Letho, et al. [14] reports 137Cs activity concentration in berries of black blueberry (V. Myrtillus) in southern Finland 1180±23 Bq kg⁻¹ dry weight (d.w.), in areas with medium Chernobyl’s fallout (about 20 kBq m⁻²); considering a mean water content of 90%, it equals approximately to 120 Bq kg⁻¹ respect to fresh weight.

For the black blueberry, soil to berry transfer factors, calculated as the ratio of fruits concentration to soil inventory (Bq kg⁻¹ dry weight)/(Bq m⁻² in the first 20 cm of soil layer) are in the range 0.05±0.07 (Table 7); however, due to the numerosity of chemical and biological processes involved, the wide variability of this data is well defined.

As 35 years from the Chernobyl accident last, the authors wonder which are the growing area of black blueberries with 137Cs activity concentrations in the range 200÷500Bq kg⁻¹. However, in the products’ label, the fruits’ geographical origin is never reported, as it is not compulsory by law.

Table 5: p-values of statistical tests performed on products and fruits (see text).

|                | 137Cs content in products | 137Cs content in fruits |
|----------------|---------------------------|-------------------------|
| Two-sample t-test | 0.35                      | 0.28                    |
| Wilcoxon rank sum test | 0.20                      | 0.15                    |

Table 6: 137Cs activity concentration in the food product based on black blueberries (canned blueberries VIS). The mass of the water measured for 137Cs content is not coincident with the mass of water used for rinsing.

| Component                  | Net weight | 137Cs (Bq kg⁻¹) |
|----------------------------|------------|-----------------|
| Canned fruit as is         | 0.05750    | 168±5           |
| Drained blueberries fruits | 0.05658    | 137±5           |
| Syrup                      | 0.06149    | 188±7           |
| Rinsing water              | 0.05306    | 5±1             |
| Drained blueberries fruits | 0.05658    | 456±12          |

(*) hypothetical original value calculated with equation 1

Table 7: Soil to blackberry transfer factors (Vaccinium myrtillus, blue blackberry).

| Species       | Transfer factors (m²/kg) | Reference                      |
|---------------|--------------------------|--------------------------------|
| V. myrtillus  | 0.0064±0.0044            | fresh weight                   |
| V. myrtillus  | 0.006                    | fresh weight                   |
| V. myrtillus  | 0.008                    | fresh weight                   |
| V. myrtillus  | 0.07*                    | fresh weight                   |
| V. myrtillus  | 0.062                    | fresh weight                   |
| V. myrtillus  | 0.052                    | fresh weight                   |

* from a single sample

Considering a mean activity concentration in black blueberries of about 100Bq kg⁻¹ (fresh weight) (the same level of the samples analysed in this paper), and considering a range of soil to berry transfer factors of 0.005±0.008m² kg⁻¹ (fresh weight), we can argue an estimate of 137Cs soil inventory of about 12.5±20Bq m⁻². As can be seen from the map in Figure 1, areas with levels of contamination between 10 and 40kBq m⁻² are very wide: these areas are found in Italy, in Scandinavia, in central Europe and in Russia. Following our hypothesis, it is reasonable to expect that on EU market black blueberries with activity concentration higher than 100Bq kg⁻¹ are in circulation and traded. Moreover, considering the data reported in Table 1, it is reasonable to assume that many lots of berries over 600Bq kg⁻¹ (maximum reference level for import in UE) could also be traded on the EU market.

Conclusions

Following the Chernobyl accident, 137Cs activity concentrations measured in food preparations based on black blueberries are to date still relevant. This is explicable observing the soil contamination spatial extension in Europe and in Russia. On the basis of the results submitted in this paper and of the maps cited above, the authors believe possible that on the EU market, fruits with 137Cs activity concentration higher than 600Bq kg⁻¹ are still introduced and in circulation.

The analysis of food preparations with organic label showed mean 137Cs activity concentrations of the same level or a little higher than those without any organic certification. On the other hand, organic labels do not comprise any radioactivity analysis, neither in raw material, nor in food products.

From the results shown in this paper, the extension of the contamination in Europe and in Russia, the wide variability...
of transfer factors, it is important that the monitoring of $^{137}$Cs content in berries continue, both with the aim to protect the population’s health, and to improve scientific knowledge.

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