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NATURAL AND ARTIFICIAL RADIONUCLIDES IN WOOD BIOMASS USED FOR HEATING – COMPARISON OF NORTH-EAST ITALY AND IMPORTED WOOD PELLETS

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Abstract. *The problem of radionuclides contamination in imported wood pellet, used for industrial and domestic heating, emerged for the first time in Italy in June 2009 in a batch from Lithuania, in which a concentration of ^{137}Cs of about 300 Bq/kg was measured, increased to about 40000 Bq/kg after burning. The radioactive fall-out due to the Chernobyl accident (26 April 1986), that deposited over extensive areas of central and northern Europe, affected also areas exploited for agroforestry and forestry resources. The soil contamination occurred almost like "leopard spot" and so in a heterogeneous way also at thousand kilometres away from the Chernobyl site. The eventual radioactive contamination currently present in woody biomasses is almost due to ^{137}Cs radionuclide, which has a half-life of about 30 years and therefore (depending on the contamination of the investigated area) still potentially present in variable quantities in soils and vegetation. The biomass represented by pellet and wood chips are nowadays of a great importance due to its extreme thermal efficiency, its cheapness and its relative low environmental impact in terms of carbon dioxide emissions; the major problem involved in this biomass, in the Italian context, is represented by the fact that the demand for pellets is much greater than the territorial possibility of self-production, therefore there is the need to import this resource from external countries and thus with potentially non-negligible levels of radioactivity. This work, conducted at ICMATE-CNR in Padua, aimed to investigate the activity concentration of ^{137}Cs, ^{40}K and other radionuclides possibly present in 27 samples of forest chips (woody flour mainly produced by Picea abies species) from the autonomous province of Trento and the ashes produced by the same samples burned in similar to domestic combustion conditions (pellet stove at about 550° C). The taken samples were subsequently analysed by high-resolution gamma spectrometry in order to evaluate the activity concentration of ^{137}Cs and of the natural ^{40}K, first in the "fresh" samples of forest chips and afterwards in the same samples incinerated after their pelletization, with the aim of comparing the amount of radionuclides of the former with respect to the latter and to determine the concentration factor. The extensive sampling to the whole province allowed to have a reliable and composite map of distribution of ^{137}Cs radioactivity in the wooded areas on the surface of the Trento province. ^{134}Cs had been also searched for, but it was below the instrumental limits of detection in all the samples. Results thus obtained were then compared and evaluated with respect both to dataset previously obtained from the same ICMATE-CNR laboratory consisting of 65 pellet samples from different areas of Eastern Europe (Bosnia, Croatia, Ukraine, Serbia and Russia) analysed in 2010-2011; and with respect to literature data relating to woody biomass for combustion of European and non-European origin.*

Keywords: *Cesium-137 in biomass, Chernobyl soil deposition, contaminated biomass, radionuclides contamination, wood pellet contamination*

1. INTRODUCTION

The use of biomass, especially woody ones, for the production of heat and electricity has become increasingly important and cheapness at global level and above all at the European level. In particular, the exploitation of biomass represented by pellets and wood chips is nowadays of great importance given its extreme thermal efficiency, cost-effectiveness and the relatively low environmental impact in terms of carbon dioxide emissions. In Italy the major problem linked to this biomass is represented by the fact that the demand for pellets is much greater than the possibility of the territory to produce it: for this reason, this product is mainly imported from foreign countries.

It is known that large regions of the Boreal Hemisphere have been subject to radionuclide depositions on the ground following accidents at nuclear plants and atomic tests in the atmosphere. While before the accident occurred at Chernobyl nuclear plant the presence of artificial radionuclides in Europe was due to radioactive fall-out following nuclear weapons testing, today it is instead mainly represented by the accident of 1986 [1].

In summary, the Chernobyl accident caused the release into the atmosphere of the most volatile fraction of the radionuclides contained in the core of the reactor and the magnitude of the event exceeded all the accidents that had previously occurred at other reactors.

In the first temporal phases after the explosion the most important radionuclides were represented by short-lived ones (mainly ^{131}I and ^{33}Xe) but while time passing their importance decreased considerably. Several months later ^{134}Cs and ^{137}Cs became the major

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source of population exposure and over time the importance of ^{137}Cs relative to ^{134}Cs grew due to its larger half-time. Today, the radionuclides most present at significant distances from the reactor are ^{137}Cs ($T_{1/2}$ 30.2 years) and ^{90}Sr ($T_{1/2}$ 28.8 years). In particular, the radioactive fall-out due to Chernobyl accident deposited in large areas of central and northern Europe [1] and also affected areas exploited for forest and agroforestry resources. It is known that the distribution of these radionuclides on the ground was not homogeneous in Europe's territory and in particular it depended on the meteorological events in the days immediately after the accident and on the amount of rainfall (see Figs. 1-3).

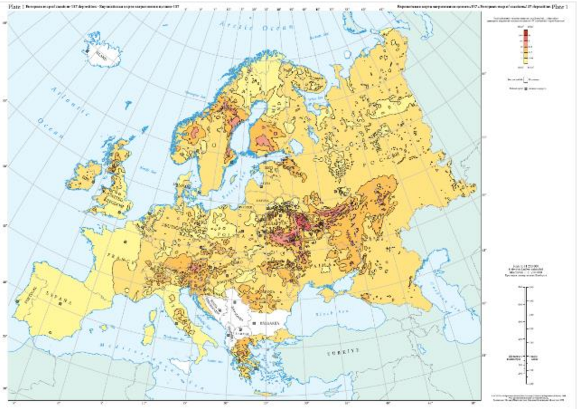


Figure 1. Map of the ground distribution of the radionuclide ^{137}Cs following the Chernobyl [1].

Figure 2. Map of the ground contamination in Italy and Croatia and a focus of the N-E zone of Italy [1].

For this reason, it is not possible to predict the concentration of artificial radionuclides in plant species on the basis of distribution maps of the surface concentration of radionuclides on the ground. In addition, the soil-to-plant transfer coefficients can be influenced by many aspects which in turn depend on variables connected to the characteristics of the soil and to any symbiotic-parasitic relationship that can develop in the root zone of trees.

In Italy the problem relating to the presence of ^{137}Cs in woody biomass emerged in June 2009 on a batch of imported pellets following a series of analyses on a low-quality product of the Lithuanian brand "Naturkraft". Once the results were obtained, the product was withdrawn from the market and seized by the Prefecture of the Republic of Aosta (territorial government office) (Tab. 1).

Table 1. Analysis carried out by the University of Pavia [2] on the Lithuanian sample.

	PELLET	ASH
	^{137}Cs (Bq/kg)	
MEAN	10.2	1530.5
MINIMUM	1.5	1192.0
MAXIMUM	20.5	1760.0
STD. DEV.	6	187

Already in 1996, some Canadian authors questioned about the possibility that fires would develop in wooded areas contaminated by radionuclides and specifically with radioactive isotopes of caesium, iodine and chlorine. It has been shown that the percentage loss of these radionuclides in the atmosphere is highly variable (from 10 to 90%) and that the solubility of these elements in the ashes decreases with increasing combustion temperature. The results report an enrichment in ^{137}Cs in the ashes from 4 to 20 times compared to the "fresh" product. Moreover, reproducing the conditions of a classic forest fire, it was seen that from 40 to 70% of Cs was dispersed and resuspended in the atmosphere (Fig. 3) [3].

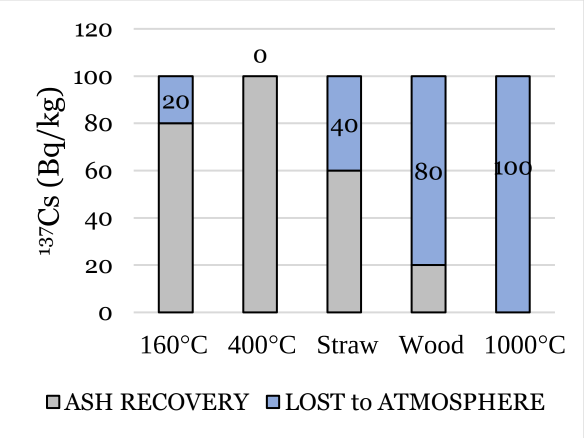


Figure 3. Percentage of ^{137}Cs recovered in ashes and lost to the atmosphere from Amiro *et al.*, 1996 [3].

Table 2. ^{137}Cs and ^{40}K in the barks of two tree species, and the ratio between ^{40}K and ^{137}Cs from Belivermiş *et al.*, 2010 [4].

	PINUS NIGRA			QUERCUS PATRAEA		
	^{137}Cs (Bq/kg)	^{40}K (Bq/kg)	^{40}K/^{137}Cs	^{137}Cs (Bq/kg)	^{40}K (Bq/kg)	^{40}K/^{137}Cs
MEAN	4.4	35.3		15.4	72.3	
MIN.	0.2	10.2		0.4	32.0	
MAX.	17.7	94.9		91.1	198.6	
STD. DEV.	4.7	18.1		22.9	31.1	
			8.1			4.7

In 2010, researchers of the University of Istanbul conducted analyses on the bark that appears to be the place of greatest accumulation of ^{137}Cs of several samples of two tree species, pine (*Pinus nigra*) and oak

(*Quercus patraea*), in order to carry out a sampling over the whole region of Thrace (Turkey) (Tab. 2). In this case the analysis was aimed at the understanding and quantitative estimation of artificial (^{137}Cs) and natural (^{40}K) radionuclides present in the area [4].

Subsequently, various Italian Customs Agencies, including those of Gorizia and Trieste, conducted radiometric analyses on 65 samples performed from August 2010 to May 2011 on over 1000 tons of pellets imported from Eastern Europe and Balkans (Fig. 4). Of the 65 samples only 6, mainly from Ukraine and one from Russia, showed values much greater than 10 Bq/kg (39.5 and 110 Bq/kg respectively) while the average of concentration was 6.8 Bq/kg for ^{137}Cs and 29.6 Bq/kg for ^{40}K [5].

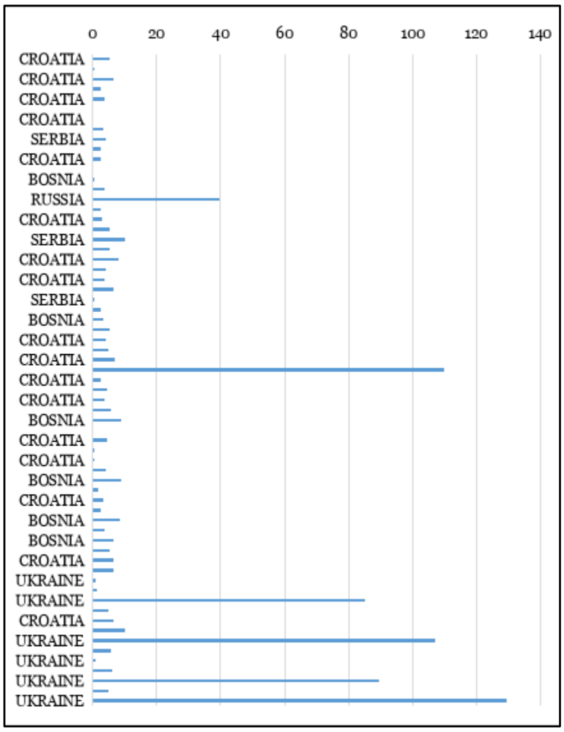


Figure 4. ^{137}Cs measured in pellet samples coming from different countries in Bq/kg [5].

With the same aim of the previous works, in 2012 some analyses were conducted on pellets imported in Italy and coming from different European and non-European countries, to evaluate the activity concentration of ^{137}Cs (Tab. 4); the results showed an activity concentration for ^{137}Cs in the pellets between 10 and 320 Bq/kg, while the ash samples of the same burnt pellet showed values of ^{137}Cs from 30 to 3176 Bq/kg; the activity concentration factor between the “fresh” product and the burnt one was between 25 and 200 [6].

Finally, between 2016 and 2017, measurements were made on wood chips and the relative ashes made from 3 samples taken from a biomass power plant in the Tarvisio area (UD). The results showed non-alarming concentrations in the pellet (from 10 to 60 Bq/kg) which, however, reached much higher concentrations in the ashes produced by combustion (about 5000 Bq/kg); some analyses were carried out also by ARPA in various regions, which showed results quite similar to the previous one, with concentrations in the pellet between 2 and 33 Bq/kg, which reach much higher concentrations in the respective ashes (between 40 and 2400 Bq/kg) [7].

Table 4. ^{137}Cs concentration in pellet samples from Desideri *et al.*, 2012 [6].

ASH	
^{137}Cs (Bq/kg)	
MEAN	571
MINIMUM	30
MAXIMUM	3176
STD. DEV.	811

The transfer factor is defined as the ratio of a radionuclide concentration in the plant mass respect to the same radionuclide concentration in the soil (TF= $\text{Bq/kg}_{\text{dry plant mass}}/\ \text{Bq/kg}_{\text{dry weight soil in the “rooting zone”}}$ [8]).

The evaluation of the soil-to-plant transfer factor of ^{137}Cs is a much-debated topic: some authors tried to propose models to estimate and evaluate the movement and the different absorption of radionuclides by the plants [8]; other authors dispute the “simplicity” of the models proposed so far, stating that the components at work in the exchange-recruitment-resuspension in the soil-to-plant system are multiple and too complicate to be expressed with a model [9].

A further influence recognized in the literature is the role of pH and the content of organic matter within the substrate: the greater absorption of caesium occurs with pH of 4-5, while at higher values it decreases. This statement is consistent with the ionic competition that would seem to exist between Cs and K, in fact the latter would be more present than Cs in contexts with higher pH, suggesting a sort of inverse proportionality relationship between the two elements in the same environment. Moreover, these statements are also in agreement with the observation that absorption of Cs is greater from soils with a high content of organic matter, and therefore basically acidic, while it is very buffered and unfavourable with respect to K in soils with a high clayey content [11].

The compartmental differentiation of the chemical elements inside the tree should also be underlined; in fact, two different accumulation zones can be distinguished: the bark and the tree. In the bark, i.e. the outermost layer, the “inert” elements not useful for the life of the tree are stored: among them there are the heavy metals and isotopes like Pb, Fe, Si, Al and Cs (if present also ^{137}Cs); instead, in the innermost part, called the “book”, all the essential elements for the plant are accumulated, including Mg, Ca, S, Zn and K (also ^{40}K) [4, 12, 14, 15]. It is therefore clear that, if the choice is to use the bark as the main constituent in the production of woody biomass, there may be a real possibility of intercepting the most contaminated component of the entire plant.

Finally, the environmental context in which the tree develops and all the exchanges that it can have with the surrounding land must be considered; the study and modelling of the root and foliar uptake of trees is an issue still under development and study. However, in addition to the different contributions of the root intake from the soil and the foliar adsorption of the atmospheric aerosol, any symbiotic relationships or parasitism in the trees itself must also be considered [16]. Today it is known that trees of almost all species coexist with colonies or single fungal individuals at their roots and this presence could influence the effective final activity concentration of radionuclides in the tree; this influence is even more complex to understand given that the horizon in which fungi tend to live is precisely the one characterized by the clayey minerals thus leading to complex dynamics of exchange and flow in the subsoil; on the other hand, in general, the presence of fungi at the roots might intercept the pollutant preventing its excessive absorption in the plant itself [17].

In this work particular attention was dedicated to the detailed analysis of the so called "concentration factor" of ^{137}Cs in the pellet defined as: the ratio of the activity concentration in the pellet samples with respect to the activity concentration in the relative ashes ($CF = Bq/kg_{^{137}Cs\ pellet} / Bq/kg_{^{137}Cs\ ash}$). This concept was used to the purpose of assessing the possible danger of the ^{137}Cs content in the ashes produced in the stove.

As regards the legislation about ^{137}Cs presence in the environment it should be considered that there is still no defined limit of radioactivity, either general or specific, on the radionuclides content in biomass; furthermore, it should be emphasized that it would be necessary not only to establish a limit for radioactivity in wood and derivatives, but also to make it restrictive for the gamma-emitting radionuclides that could be more present, and specifically ^{137}Cs and ^{40}K. In addition, attention is to be paid to the importation of biomasses from potentially contaminated areas and above all, pellets that have the raw materials made from the bark, as this is a bioaccumulation district of minerals and metals and so maybe also of the ^{137}Cs radionuclide [5].

A study was conducted in 2022 in the ICMATE-CNR radiochemistry laboratory on 27 pellet samples collected from the autonomous province of Trento, which were analysed as "as it is" (= pelletized product such as that available on the market) and in their burnt ash form; the combustion, as already mentioned, was carried out using a pellet stove available on the market and so used for domestic purposes; the aim was to make the analysis as faithful as possible to the conditions that could arise in any domestic context. In fact, this work for the first-time deals with the recreation of the conditions of burning in a domestic stove more similar to those ones of the potential users of the product while, previously, the tests and measurements had been made with laboratory stoves and muffles.

2. MATERIALS AND METHODS

2.1. Sample preparation

The sampling took place during May 2022 and involved the whole province of Trento, divided previously into macro-areas, and consisted in the withdrawal of 60 litres of wood chips for each point in the map (Fig. 5), for a total of 27 samples.

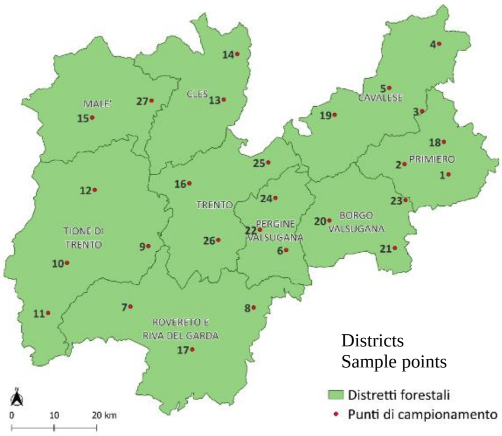


Figure 5. Sampling points and forest districts.

The sampling project foresaw that the wood chips were mainly produced from the arboreal species of spruce (*Picea abies*); the wood chips were produced directly in the forest after the trees were felled. Five samples are composed of mixed species (*Picea abies, Pinus sylvester* and *Pinus nigra*). Afterwards in laboratory the wood chips were pelletised using a special instrumentation (Analysis Lab. BioFuels Dep. TESAF – Unipd – Legnaro (PD) – ITALY) in order to burn the samples in a domestic-like stove in conditions that reproduce the domestic standard at the best. After drying the samples of wood chips, these was grounded up until obtaining a wood sawdust which was finally compacted with a pellet mill until obtaining 3.15 mm pellet shavings. The objective of these preparations was to create the pellet's ashes (from the wood chips) which would allow an analysis of the quantity of radionuclides and their possible concentration between the "fresh" sample and its ashes. In the laboratory, the ashes were obtained from the combustion of 8 kg of pellets for each sample in a domestic stove at a temperature of 550°C [19].

2.2. Gamma spectrometry measurements

The measurements with gamma spectrometry were made with high-resolution hyper-pure germanium detectors (HPGe) in the ICMATE-CNR physical laboratory; each detector is placed in a 100 mm thick lead well, in order to shield it from the environmental background; additional inner sheets of Cd and Cu provide a shielding from low energy photons (mainly "secondary").

The "as it is" pellet samples were analysed in the so called "Marinelli" containers of 1 litre, while the ash samples were measured in 50 ml polystyrene jars. The efficiency calibration for this sample volume configuration has been performed with multipeak certified standard solutions counting QCY48 and QCYB40 by Amersham. The acquisition time was different for each sample and was chosen with the aim of achieving a statistical counts error less than the 10%

of the photopeaks of interest. Spectra were acquired with GENIE-2000 software (® AREVA/ Canberra).

3. RESULTS AND DISCUSSION

In tables 5 and 6, the main descriptive statistics of ^{137}Cs and ^{40}K concentration in our samples are reported. In Table 7, our results about ^{40}K and ^{137}Cs concentration are compared with literature data.

The mean value of activity concentration of ^{137}Cs in pellets from Trentino region is quite low (4.7 Bq/kg) respect to other authors [2,4,5]. In fact, as considered in the introduction, pellets from eastern European countries might have ^{137}Cs activity concentration also higher than 100 Bq/kg. For this reason, also ^{137}Cs concentration in ash is less than data reported by [2, 6]. On the contrary ^{40}K activity concentrations of our samples are quite similar to other regions [4].

We studied the potential relationship between ^{137}Cs and ^{40}K concentration in wood (in pellet samples), investigating the possible variation of ^{137}Cs uptake depending on K concentration in the soil (and so of ^{40}K), as discussed in the introduction. In Fig. 6, ^{137}Cs concentration in pellet samples is plotted vs ^{40}K conc.; in Fig. 7, ^{137}Cs concentration in ash samples is plotted vs ^{40}K.

We do not appreciate any correlation, as can be seen also by the trend line. We have to notice that the ^{40}K concentration range is quite small in our samples, so it would be unlikely to appreciate any relationship. Also, ^{137}Cs concentration range in soils in the area investigated from us is very small. The same consideration is valid also for ^{137}Cs and ^{40}K concentration in ash samples.

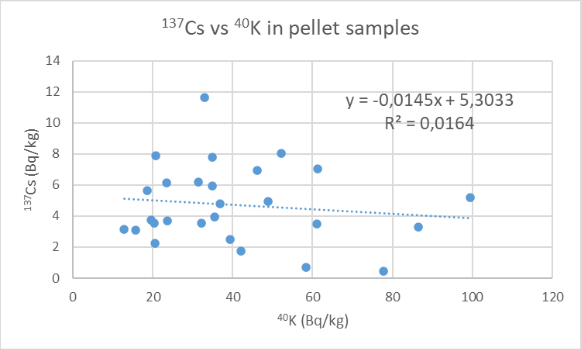


Figure 6. Ratio scatterplot of ^{40}K and ^{137}Cs in ours pellet samples.

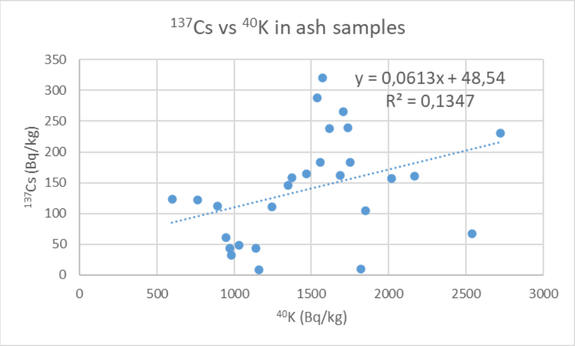


Figure 7. Ratio scatterplot of ^{40}K and ^{137}Cs in our ash samples.

Table 5. Our results obtained from pellet samples.

PELLET			
	^{137}Cs (Bq/kg)	^{40}K (Bq/kg)	^{40}K/^{137}Cs
MEAN	4.7	40.3	18.2
MINIMUM	0.4	12.8	2.6
MAXIMUM	11.7	99.5	182.0
STD. DEV.	2.5	21.8	35.7

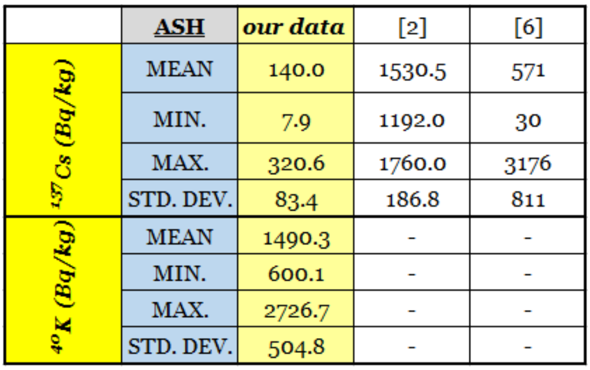
Table 6. Our results obtained from ash samples.

ASH			
	^{137}Cs (Bq/kg)	^{40}K (Bq/kg)	^{40}K/^{137}Cs
MEAN	140.0	1490	25.1
MINIMUM	7.9	600	4.9
MAXIMUM	320.6	2726.7	203.2
STD. DEV.	84.4	504.8	43.9

Table 7. Comparison between literature data and those obtained in this work about ^{137}Cs and ^{40}K in pellet.

	PELLET	***our data***	[2]	[4]	[5]
^{137}Cs (Bq/kg)	MEAN	4.7	10.2	10.8	13.2
	MIN.	0.4	1.5	0.2	0.6
	MAX.	11.7	20.5	91.1	129.3
	STD. DEV.	2.5	6.4	18.0	28.0
^{40}K (Bq/kg)	MEAN	40.3	-	55.3	-
	MIN.	12.8	-	10.2	-
	MAX.	99.5	-	198.6	-
	STD. DEV.	21.5	-	31.8	-

Table 8. Comparison between literature data and those obtained in this work about ^{137}Cs and ^{40}K in ash.



	ASH	***our data***	[2]	[6]
^{137}Cs (Bq/kg)	MEAN	140.0	1530.5	571
	MIN.	7.9	1192.0	30
	MAX.	320.6	1760.0	3176
	STD. DEV.	83.4	186.8	811
^{40}K (Bq/kg)	MEAN	1490.3	-	-
	MIN.	600.1	-	-
	MAX.	2726.7	-	-
	STD. DEV.	504.8	-	-

4. CONCLUSION

- The ^{137}Cs content in the woody biomass produced in Europe is highly variable and cannot be predicted «a priori» due to all the mentioned factors.
- It can be assumed that the highest concentrations of ^{137}Cs can be found in samples from highly contaminated areas and will also be higher if it is used the bark of the tree.
- ^{40}K is always present in woody biomass and is concentrated quite significantly in the ashes.
- We suppose that if a bark from a contaminated area is used, there will be a good chance of being in the worst-case condition regarding the activity concentration of ^{137}Cs.
- Currently, there is no regulatory limit on the content of radionuclides in biomass for combustion and we believe that it should be set individually for each single radionuclide.
- In order to propose an indicative level of the radionuclide concentration, which might indicate the quality of the pellet itself, it is important to also take into consideration the natural radionuclides present, among which the most abundant is ^{40}K.

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