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Quality Characteristics and Radioactive Contamination of Wood Pellet Imported in Italy

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Abstract

The problem of Caesium-137 (137 Cs) contamination of the imported wood pellet used for burning has been reported in Italy since June 2009. Since then, sampling and analysis were performed at the crossing border points of the provinces of Trieste and Gorizia, on request of the Health and Customs Border Bureau. This paper presents the results of the analysis performed on 65 samples from August 2010 to March 2012, which covered a total of products over 1500 tons of various origins, imported from Eastern Europe and the Balkans. Most of the samples showed very low 137 Cs activity concentrations; only a few hot spots showed 137 Cs activity concentrations higher than 100 Bq·kg^{-1}. The results of dose evaluations for wood pellet stoves users under the hypotheses assumed in this study were largely below the threshold of radiological relevance.

Keywords

Radioactivity, Wood Pellet, Biofuels, Caesium-137, Radioprotection

1. Introduction

At present, the conglomerate wood pellet represents one of the most used biomass heat sources. It is obtained through simple mechanical processes, during which very fine sawdust is exposed to high pressures and contemporaneously is passed through a spinner with 6 - 12 mm holes. The heat produced during these steps activates the lignin, a substance naturally present in the wood, bringing into operation its binding property that determines

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the characteristic compactness and cylindrical shape of the product. The wood pellet represents a valid alternative to traditional heating fuels, because it is easily available, practical to transport, ecological and safe. It is usually packed in 15 kg bags, not cumbersome and easy to move. The main characteristics of wood pellet (see **Table 1**) are the high thermal efficiency (over 80%) and the very low amount of residual ashes (0.3% - 0.6%). The demand of wood pellet constantly rose during the last years, causing an obvious remarkable increase of its price, even if it is still competitive with respect to traditional fossil fuels. The thermal efficiency of this material depends on the quality of wood used for its production. Generally, the best wood pellet is obtained from fir or beech wood; it must have a light or "white" colour, and the label on the package must report its main characteristics (thermal efficiency, place of origin, etc.) and quality references according to international regulations such as DIN 51731 or Ö-NORM M7135 [1]. Also, it must have regular shape and dimensions, corresponding to those reported on packages; it must not have excessive amounts of residual sawdust, and it must be smooth, compact and shiny.

There are many different good quality wood pellets, even if the darkest ones should be evaluated more carefully. About the latter, it must be considered that wood pellet stoves are adjusted to be used with the most widespread wood pellets (light colored, produced from fir or beech wood); the use of other wood pellets may cause malfunctions and lower performances, so an appropriate adjustment of burners should be necessary. Generally, wood pellet is mainly produced in countries with large woody surfaces available (such as Russia, Canada, Sweden, Germany, Austria, Switzerland and the Baltic countries). Italy is a large consumer of wood pellet, but it is not a great producer (there are only some small facilities in north-eastern and in central regions), and then wood pellets for sale are almost exclusively imported. Instead, Italy is a large producer of wood pellet-based heating systems (stoves, boilers, fireplaces) and it is a large market for them [2].

2. Italian and European Legislation References

At present in Italy, such as in the other UE countries, there is a lack of legislation protecting consumers about the wood pellet quality. The main European requirements for wood pellet are listed in **Table 2**; however, a specific limit for Caesium-137 (137 Cs) is not established.

Some Italian national producers, united in the association AIEL ("Associazione Italiana Energie Agroforestali", Italian Agroforestry Energy Association), voluntarily trusted the quality of their product to the trademark "Pellet Gold", which is based on the standard UNI/TS 11263:2007 [3]. For some aspects, this trademark [4] is more restrictive than the cited UNI/TS rule; for example, an additional parameter, here introduced and not present in any other certification requirement, is the formaldehyde content, which is fundamental to notify the presence of potentially harmful chemicals, such as glues and paints. A limit of 6 $Bq \cdot kg^{-1}$ for the overall radioactivity of the product is also established, but no explanation about such a value is given, neither which radionuclides are to be monitored.

3. The Radioactive Contamination of Wood Pellet

The problem of radioactive contamination of wood pellet arose in Italy in June 2009 on an imported product, after a series of analysis required by consumers unsatisfied with the quality of the purchased goods.

The "Naturkraft" brand wood pellet, imported from Lithuania, was casually found to be contaminated by 137 Cs; for this reason the local authority "Pretura" (magistrate's court) in Aosta ordered to confiscate it over the

Table 1. Main technical characteristics of a good quality wood pellet [2].

Characteristic	Typical value
Diameter	6 - 12 mm
Lenght	15 - 50 mm
Density	1150 - 1400 $kg \cdot m^{-3}$
Humidity	8% - 12%
Thermal power	4.7 - 5.5 $kW \cdot kg^{-3}$
Residual ashes	0.3% - 0.6%

Table 2. European requirements for wood pellet.

Specifications	Units	CEN/TS[a]	Austria	Germany	Sweden[b]	PFI (US)
Gross calorific value	MJ/kg	20.00 - 20.47	>18.00	17.45 - 19.54	>16.91	18.61
Ash	%, dw	0.3	<0.5	<1.5	≤0.7	<1
Moisture	%, as received	<10	<12	<12	≤10	<8
Chlorides	ppm	<100 - 300	<200	<300	≤300	<300
Sulphur	%	<0.01 - 0.005	<0.04	0.08	≤0.08	
Nitrogen	%	<0.1 - 0.5	<0.30	0.3		
Arsenic	mg/kg			0.8		
Cadmium	mg/kg	0.1		0.5		
Chromium	mg/kg	1		8		
Copper	mg/kg	2		5		
Mercury	mg/kg			0.05		
Lead	mg/kg			<10		
Zinc	mg/kg	10		<100		

[a]European committee for standardization as given in CEN/TS 14961 "Annex A" examples of specifications for high quality classes of solid biofuels recommended for household usage; [b]Sweden Group 1 (best) specification [5].

entire national territory, for a total amount of 10,000 tons. After this event, some initiatives were undertaken by many organisations to evaluate the real risk for public health and to propose limits, if necessary. In this case, the Agency for Customs ordered radiometric controls on imported stocks similarly to other goods. It must be observed that wood pellet is not subject by current laws to any radiometric limit in all the EU countries. In fact the activity concentration limit of 1 $Bq \cdot kg^{-1}$ provided for by the Italian regulation, legislative decree 230/95 (D. Lgs, 1995) is referred only to practices with radioactive materials; so, in this case, it must be intended only as a reference value, as burning wood pellet cannot be considered a practice with radioactive materials. The problem of the 137 Cs content of wood pellet arises from the fact that this product is imported to be used as a fuel in heating systems. The combustion causes a concentration of mineral salts, 137 Cs among them, roughly of a factor 100 - 200 and the ashes derived from also slightly contaminated wood pellet could therefore have an high 137 Cs activity concentration and could be potentially an health hazard. Radioprotection problems may arise in the management of wood pellet ashes during the cleaning operations of the domestic stoves, because they are a waste containing 137 Cs, even if, as above reported, they cannot be considered a "radioactive waste" as described by the legislative decree 230/95 [6]. From an environmental point of view, the most relevant worry is due to the use of the ashes as fertilizers for orchards and gardens. It must be noted that the health risk for population was initially evaluated as "non-significant" by the ARPA ("Agenzia Regionale per la Protezione Ambientale", Regional Agency for Environmental Protection) of region Piedmont [7]. In fact, the dose[1] value for the population was estimated very low, far from the levels of radiological concern (10 $\mu Sv \cdot year^{-1}$), even if authors themselves reserved to update their evaluation when further data would become available. Shortly afterwards, a study of University of Pavia found a similar conclusion [8].

The Lithuanian "Naturkraft" wood pellet brand was partially released from attachment in September, 2009, except for the batch sequestrated in Region Valle d'Aosta. Since then, the competent Customs Offices continued to require a certificate of radiological conformity for imported batches through gamma-rays spectrometry analysis, to measure the amount of the potential contamination by 137 Cs. These analyses were performed even in absence of a limit for the considered material, as at present there are no limits for this good. This situation seems to be almost partially due to ISPRA ("Istituto Superiore per la Protezione e la Ricerca Ambientale", National Institute for Environmental Protection and Research) doubts about the effect of accumulation of 137 Cs in ashes,

[1]Radiation protection quantity that indicates the amount of energy for unit mass absorbed by an organism exposed to ionizing radiations; it is used also for estimating the risk of long term stochastic health effects following radiation exposure.

which could spread in the environment in an uncontrolled manner, and accumulate in the food chain. In fact, if the amount of wood pellet imported in Italy is considered, a hypothetical contamination of only a few $Bq \cdot kg^{-1}$ would imply the introduction in the Italian boundaries of some thousands of MBq of 137 Cs for year. This amount, even if diluted in a very large bulk, can give a considerable worry about the doses that could be received by particular groups of the population. The topic addressed in this paper results from the need of estimating the amount of 137 Cs in the imported wood pellet used as heating fuel.

4. Materials and Methods

From August, 2010 until March, 2012, during the radiological survey carried out in the customs area of Trieste and Gorizia, and co-ordinated with the local Agency for Customs, 65 samples of wood pellet were collected and analysed, each one derived from a single bulk. The determination of the 137 Cs and the Potassium-40 (40 K) activity concentration was performed through high resolution gamma spectrometry. The instrument used is based on an intrinsic hyper-pure Germanium detector with Be window; the major characteristics of the detector are listed in **Table 3**. The efficiency and energy calibrations were obtained by means of certified gamma reference standards of mixed radionuclides [9] [10]. Samples were taken, for each charge, from three different bags near the back door of the truck. Afterwards the samples coming from each charge were mixed in order to obtain the average sample for the analysis. The untreated average sample were put each one in 1 L Marinelli beaker; all the measurements were performed with a low background configuration.

5. Results and Discussion

The main data regarding the analysed wood pellet samples and the results of gamma spectrometry analysis are reported in **Table 4**. The 40 K activity concentrations found and the data spread are in agreement with 40 K contents normally found in wood. Also 137 Cs is found in wood as a result of its presence in the environment, in consequence of weapon tests and of Chernobyl accident. 134 Cs (which has the same origin of 137 Cs) in all the analysed samples was always below the detection limits; anyway, it must be noted that its half-life time (2.07 years) is shorter than 137 Cs (30.17 years), so it underwent a larger decay. The total imported wood pellet amount in the considered period was 1550 tons, with an average activity of 13.2 $Bq \cdot kg^{-1}$ for 137 Cs, and of 43.0 $Bq \cdot kg^{-1}$ for 40 K; then, the theoretical total amount of imported 137 Cs should be 20.46 MBq. As reported in **Table 4**, the highest values were found in three samples coming from Ukraine and exceeding 100 $Bq \cdot kg^{-1}$; in all the other samples 137 Cs was generally less than 10 $Bq \cdot kg^{-1}$. Considering the quality and healthy of wood pellet, especially from a radioprotection aspect, it should be important to avoid the importation of products made with the cortex of trees, which is responsible for the bio-accumulation of mineral salts and metals and then also of most of 137 Cs [11] [12]. This kind of product can be generally distinguished by its dark brown colour; for this reason, clear wood pellet should be preferred.

Table 3. Main characteristics of the germanium detector used.

Characteristic	Typical value
Detector type	Coaxial, N-type, with Be window
Constructor	Canberra
Diameter	53.5 mm
Length	53.0 mm
Distance between Ge detector and Be window	5 mm
FWHM (Full Width at Half Maximum) (at 122 keV)	0.81 keV
FWHM (at 1332 keV)	1.73 keV
Peak/Compton	55.9/1 (at 1332 keV)
Relative efficiency	25.3% (at 1332 keV)
Depletion voltage	−2500 V
Recommended voltage	−4000 V

Table 4. Collected samples.

No.	Date	Origin	Amount (tons)	Customs	^{137}Cs (Bq·kg^{-1})	^{40}K (Bq·kg^{-1})
1	17.08.2010	Croatia	24.150	GO	5.4	58.1
2	17.08.2010	Croatia	24.882	GO	0.6	38.0
3	23.08.2010	Croatia	24.625	GO	6.5	29.0
4	03.09.2010	Croatia	24.150	GO	2.5	50.3
5	28.09.2010	Croatia	25.200	TS	3.7	47.8
6	29.09.2010	Croatia	25.200	TS	<0.5	73.3
7	06.10.2010	Croatia	24.400	TS	<0.5	46.9
8	08.10.2010	Croatia	24.495	Mn	3.5	53.0
9	12.10.2010	Serbia	24.500	GO	4.3	52.0
10	12.10.2010	Croatia	25.200	TS	2.4	32.8
11	14.10.2010	Croatia	25.200	TS	2.5	35.8
12	28.10.2010	Croatia	25.200	TS	<0.5	28.0
13	29.10.2010	Bosnia	24.360	TS	0.7	42.4
14	04.11.2010	Bosnia	24.600	TS	3.9	67.1
15	04.11.2010	Russia	22.000	GO	39.5	51.0
16	05.11.2010	Croatia	25.200	GO	2.5	23.7
17	11.11.2010	Croatia	16.600	GO	2.8	52.0
18	15.11.2010	Bosnia	24.600	TS	5.5	41.8
19	15.11.2010	Serbia	24.500	GO	9.9	59.0
20	25.11.2010	Bosnia	24.600	TS	5.5	31.0
21	30.11.2010	Croatia	25.880	GO	7.9	44.0
22	09.12.2010	Croatia	25.200	GO	4.2	29.0
23	13.12.2010	Croatia	24.720	GO	3.6	36.8
24	14.12.2010	Croatia	25.200	TS	6.7	48.5
25	16.12.2010	Serbia	24.500	GO	0.7	23.0
26	17.12.2010	Croatia	22.950	GO	2.5	36.6
27	22.12.2010	Bosnia	24.360	TS	3.4	34.7
28	23.12.2010	Croatia	22.400	GO	5.5	62.0
29	10.01.2011	Croatia	25.200	TS	4.3	29.6
30	13.01.2011	Croatia	23.058	Mn	5.0	47.2
31	13.01.2011	Croatia	23.600	TS	7.0	38.9
32	19.01.2011	Ukraine	22.000	GO	110.0	35.5
33	27.01.2011	Croatia	25.200	Mn	2.5	21.8
34	27.01.2011	Croatia	23.600	TS	4.4	26.0

Continued

35	27.01.2011	Croatia	23.600	TS	3.7	50.2
36	28.01.2011	Bosnia	24.480	TS	5.8	28.4
37	04.02.2011	Bosnia	24.480	TS	9.0	43.5
38	07.02.2011	Ukraine	20.650	TS	<0.5	71.6
39	10.02.2011	Croatia	24.495	Mn	4.5	42.1
40	12.02.2011	Ukraine	19.800	TS	0.6	49.3
41	15.02.2011	Croatia	25.200	Mn	0.6	66.0
42	17.02.2011	Croatia	23.058	Mn	4.2	47.3
43	24.02.2011	Bosnia	24.360	TS	8.7	39.0
44	14.03.2011	Croatia	24.030	Mn	1.7	29.6
45	25.03.2011	Croatia	23.935	Mn	3.2	25.5
46	28.03.2011	Bosnia	24.000	Mn	2.5	24.0
47	26.04.2011	Bosnia	24.000	Mn	8.3	45.7
48	06.05.2011	Croatia	25.200	Mn	3.7	39.0
49	11.05.2011	Bosnia	24.360	TS	6.5	44.4
50	23.05.2011	Croatia	24.350	Mn	5.5	43.2
51	21.10.2011	Croatia	25.000	TS	6.6	39.0
52	24.10.2011	Bosnia	24.480	TS	6.7	58.3
53	14.11.2011	Ukraine	21.600	TS	0.9	27.0
54	18.11.2011	Ukraine	21.600	TS	1.5	36.9
55	29.11.2011	Ukraine	22.044	TS	85.3	72.0
56	11.01.2012	Bosnia	24.480	TS	5.1	67.7
57	13.01.2012	Croatia	25.000	TS	6.6	25.4
58	16.01.2012	Ukraine	21.966	TS	10.0	47.7
59	16.01.2012	Ukraine	21.044	TS	107.1	54.0
60	19.01.2012	Bosnia	24.480	TS	5.6	38.6
61	31.01.2012	Ukraine	22.250	TS	0.9	58.0
62	3.02.2012	Croatia	25.000	TS	6.2	50.5
63	13.02.2012	Ukraine	22.044	TS	89.4	41.0
64	17.02.2012	Croatia	25.000	TS	5.1	27.8
65	15.03.2012	Ukraine	22.044	TS	129.3	34.6
		MEAN			13.2	43.0
		Standard deviation			28.26	13.3
		Minimum value			0.6	21.8
		Maximum value			129.3	73.3

TS = Trieste trucks terminal-industrial harbour, Monrupino; GO = S. Andrea trucks terminal, Gorizia; Mn = Monfalcone harbour.

As far as radioprotection is concerned, the main worry regards 137 Cs concentration in ashes, as concentration factors ashes/pellets of 100 - 200 are reported. If ashes have a high 137 Cs activity, the periodic cleaning of stoves from ashes may cause external irradiation dose and potentially also inhalation dose due to resuspended ashes. It is possible to estimate the dose from irradiation and inhalation considering different hypothetical scenarios:

a) a flat (about 70 - 80 m^2) in Central Italy using a wood pellet stove only for heating, consuming about 1620 kg of wood pellets for year (small consumer);
b) a single house (more than 150 m^2) in Northern Italy using the pellet stove also for hot water production, consuming about 6376 kg of wood pellets for year (large consumer);
c) a central heating system for a condominium of 10 flats, consuming about 15,000 kg of wood pellets per year.

The external irradiation absorbed dose received over the course of one year was estimated for the three scenarios above (**Table 5**), on the basis of the maximum 137 Cs activity concentration in wood pellets found in this paper (110 $Bq \cdot kg^{-1}$). Cases b) and c) consider critical groups of the population from radioprotection point of view, since the calculations regard the most exposed people (*i.e.* those which periodically clean the stove) in cases of high 137 Cs activity concentration and a strong use of wood pellet stoves. The inhalation absorbed dose for the three scenarios was calculated in **Table 6**. From the results of **Table 5** and **Table 6** the prevailing dose contribution is due to inhalation. The contribution, in any case very low, can be also lower if the periodic cleaning of the stove is done with vacuum cleaners instead of by hand. The choice of a reference level for the 137 Cs

Table 5. Irradiation dose estimation for three scenarios.

	Flat in Central Italy	Single House in Northern Italy	Condominium Stove
Mass of wood pellets burned in one year ($kg \cdot year^{-1}$)	1620	6376	15,000
Concentration factor pellets/ashes	150	150	150
Mass of ashes produced in one year ($kg \cdot year^{-1}$)	10.8	42.5	100
^{137}Cs activity concentration in pellets ($Bq \cdot kg^{-1}$)	110	110	110
^{137}Cs activity concentration in ashes ($Bq \cdot kg^{-1}$)	16,500	16,500	16,500
Mass of ashes removed for each cleaning operation (kg)	1	1.42	4
Maintenance operation duration (minutes)	10	10	60
Number of maintenance operations repetitions	11	30	25
Mean distance from ashes (m)	0.4	0.4	0.6
Dose rate at 1 m distance for 1 MBq of ^{137}Cs ($nSv \cdot h^{-1}$)	75	75	75
Dose rate at operation distance for 1 MBq of ^{137}Cs ($nSv \cdot h^{-1}$)	468.8	468.8	208.3
Irradiation dose for one year (nSv)	14.2	54.9	343.8

Table 6. Inhalation dose estimation for three scenarios.

	Flat in Central Italy	Single House in Northern Italy	Condominium Stove
Concentration of the dust resuspended in air ($\mu g \cdot m^{-3}$)	50	50	50
Breathing rate ($m^3 \cdot hour^{-1}$)	1.2	1.2	1.2
Maintenance operation duration (minutes)	10	10	60
^{137}Cs activity concentration in ashes ($Bq \cdot kg^{-1}$)	16,500	16,500	16,500
Inhalation dose coefficient for ^{137}Cs ($Sv \cdot Bq^{-1}$)	3.9E−05	3.9 E−05	3.9E−05
Number of maintenance operation repetitions	11	30	25
Inhalation dose for one year (nSv)	70.8	193.1	965.3

content in wood pellets should result from the calculated absorbed dose by critical groups of the population as far as wood pellets use is concerned. However, the reaching of an inhalation dose of 10 μSv·year^{-1} (under this threshold, the dose is considered as "not relevant" from a radioprotection point of view) would require a 137 Cs activity concentration in wood pellet of about 1200 Bq·kg^{-1}, never found in Italy so far.

6. Conclusions

Most of the imported wood pellet samples showed very low 137 Cs activity concentrations; only a few hot spots showed 137 Cs activity concentrations higher than 100 Bq·kg^{-1}. This fact and the possibility that radionuclides accumulation in ashes could interest directly critical groups of the population, the food chain and also the environment, can justify the continuation of the current control procedures. However, the dose evaluations for wood pellet stoves users under the hypotheses assumed in this paper were largely below the threshold of radiological relevance.

In addition, the unavoidable presence in wood of natural radionuclides should be taken in account in order to establish reasonable limits for radionuclides content in wood pellet. In fact, the assigned limit of 6 Bq·kg^{-1} for the overall radioactivity of the product [4] seems to be illogical: for example, in this study a mean value for 40 K (a natural radionuclide) activity concentration was found to be 43.0 Bq·kg^{-1}.

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