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Short communication

^{137}Cs IN SOIL AND FALLOUT AROUND ZAGREB (CROATIA) AT THE TIME OF THE FUKUSHIMA ACCIDENT

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This paper addresses the noticeable increase of ^{137}Cs activity concentrations in soil and fallout in the area surrounding Zagreb (Croatia) that occurred at the time of the 2011 Fukushima accident. This topic is important for public health as ^{137}Cs is highly toxic due to its long half-life of radioactive decay and chemical similarity to potassium. ^{137}Cs concentrations in fallout were much greater than in soil, but remained present longer in the latter. While being detectable in our measurements, ^{137}Cs did not spread through the food chain in amounts exceeding the maximum allowed level of radioactive food contamination. However, more thorough and consistent measurements need to be done in order to establish the precise activity trends of ^{137}Cs in Zagreb soil and fallout.

KEY WORDS: *gamma-ray spectrometry*, *public health, radioactive contamination*

^{137}Cs is one of the most abundant radioactive products of uranium fission in nuclear power plants and weapons. Therefore, ^{137}Cs calls for constant awareness, since it may pose a serious threat to human health for two main reasons. The first reason lies in the general chemical similarity between caesium (Cs) and potassium (K), which means that Cs can replace K in its biochemical role within the human body. This replacement may in turn cause strong toxic effects due to the ionisation processes that originate from radioactive decay in living tissue. The second reason is that ^{137}Cs has a rather long half-life of 30.1 years, which prolongs toxic effects over a period considerably lengthy for an average human lifetime.

When the human body is exposed to ^{137}Cs from the environment, it is affected by the emission of 661.62 keV gamma rays, while the simultaneous emission of beta particles is less serious because skin acts to protect the body. Once ^{137}Cs enters the body, skin no longer protects it and beta particles pose an equally dangerous threat as gamma rays. That is why ^{137}Cs is a severely toxic substance, especially if it enters the body via inhalation or food.

It is very important to continuously monitor the presence of ^{137}Cs in soil and fallout, as the food chain inevitably involves these pathways. This is particularly imperative in cases of nuclear accidents such as the one that occurred at the Fukushima I Nuclear Power Plant, Japan on 11 March 2011. Even though the city of Zagreb and Fukushima are considerably distant, a related temporary increase of the ^{137}Cs activity in air was observed (1). Fortunately, it remained well below a value that should cause concern. In this paper, we present the results of ^{137}Cs activity at the time of the Fukushima accident in soil and fallout sampled in the area surrounding Zagreb as a starting point for future, more detailed, measurements and analyses.

MATERIALS AND METHODS

As part of our regular measurements each spring, we sampled uncultivated soil at a location at the eastern outskirts of Zagreb. This was carried out using a corer of 10 cm in diameter to sample soil layers corresponding to depths of 0 cm to 5 cm, 5 cm to 10 cm, and 10 cm to 15 cm. The collected soil was dried, sieved, heated at 450 °C to a constant weight, homogenised (2), and finally put into Marinelli beakers (1 L in volume) to form samples for gamma-ray spectrometry.

Fallout was collected continuously throughout 2011 at the Institute for Medical Research and Occupational Health in Zagreb, using a funnel with a 1 m^2 catchment area placed 1 m above ground. Fallout from a three-month collection period was treated as a single sample and its volume was reduced to 1 L by evaporation.

Measurements of activity were carried out by high-resolution gamma-ray spectrometry using a method accredited according to the HRN EN ISO/IEC 17025:2007 standard. We used a high-purity Ge photon detector system ORTEC HP GMX of a 74 % relative efficiency and resolution of 2.26 MeV, all at 1.33 MeV ^{60}Co.

The calibration of the measurement setup was carried out using standards prepared by the Czech Metrology Institute, whereas quality assurance was performed through regular participations in interlaboratory comparisons organised by the International Atomic Energy Agency (IAEA), World Health Organisation (WHO), and European Union Joint Research Centre (JRC) (3). Soil samples were counted for a minimum of 80,000 s, and fallout samples for a minimum of 250,000 s.

RESULTS AND DISCUSSION

The appearance of excess ^{137}Cs in soil and fallout from the area surrounding Zagreb was a topic particularly worthy of investigation after the unfortunate occurrence of the Fukushima accident. We compared the values of ^{137}Cs activity concentrations (A) obtained between January 2007 and March 2011 with those measured between March 2011 and December 2012, which is also planned for future periods.

In Table 1, A (together with corresponding uncertainties) in three adjacent layers of soil is presented.

Activity in the absence of accidental conditions, denoted in Table 1 as "average 2007 to 2010", was calculated by averaging data for the period between 2007 and 2010. We used weighted statistics in which the statistical weight of an nth contribution $A_n \pm \sigma_n$ equalled $1/\sigma_n^2$ (4). Hence, the average $\overline{A}_{\text{soil, 07–10}}$ obtained was given by

$$\overline{A}_{\text{soil,07-10}} = \frac{\frac{A_{07}}{\sigma_{07}^2} + \frac{A_{08}}{\sigma_{08}^2} + \frac{A_{09}}{\sigma_{09}^2} + \frac{A_{10}}{\sigma_{10}^2}}{\frac{1}{\sigma_{07}^2} + \frac{1}{\sigma_{08}^2} + \frac{1}{\sigma_{09}^2} + \frac{1}{\sigma_{10}^2}}, \quad (1)$$

and its uncertainty by

$$\sigma_{\text{soil,07–10}} = \left(\frac{1}{\sigma_{07}^2} + \frac{1}{\sigma_{08}^2} + \frac{1}{\sigma_{09}^2} + \frac{1}{\sigma_{10}^2} \right)^{-1/2} \quad (2)$$

The subscript in a summand denotes the year (e. g., subscript 08 implies the year 2008). It is obvious that A increased in 2011 for all layers by 18 % to 60 % above $\overline{A}_{\text{soil, 07–10}}$, which was followed by a moderate decrease in 2012. Only in the deepest layer did A return to a value close to $\overline{A}_{\text{soil, 07–10}}$ in 2012, whereas it remained higher for the upper two layers.

In Figure 1, the temporal evolution of A measured in the fallout is shown. Before the accident, A showed certain variations but remained below 4 Bq m^{-3}. In the first quarter of 2011, A increased strongly, reaching A_{max} =(15.3±0.5) Bq m^{-3}. It should be noted that the fallout sample for this paper was collected between 1 January and 31 March 2011, which resulted in the fact that 57 % of the sample referred to the period after the accident. It was therefore likely that A corresponding to the period between 11 March and 31 March was even larger than A_{max}.

Table 1 *Yearly values (in spring) of ^{137}Cs activity concentrations in three layers of soil in the period 2007-2012.*

	A / Bq kg^{-1}						
Depth / cm	**2007**	**2008**	**2009**	**2010**	**average 2007 to 2010**	**2011**	**2012**
0 to 5	16.4±0.5	17.2±0.4	16.4±0.4	14.4±0.4	16.1±0.2	20.4±0.2	18.5±0.3
5 to 10	18.3±0.4	12.2±0.4	12.3±0.4	14.2±0.4	14.3±0.2	22.9±0.3	17.8±0.2
10 to 15	17.1±0.4	23.5±0.5	16.1±0.4	15.0±0.4	17.4±0.2	20.5±0.2	17.5±0.2

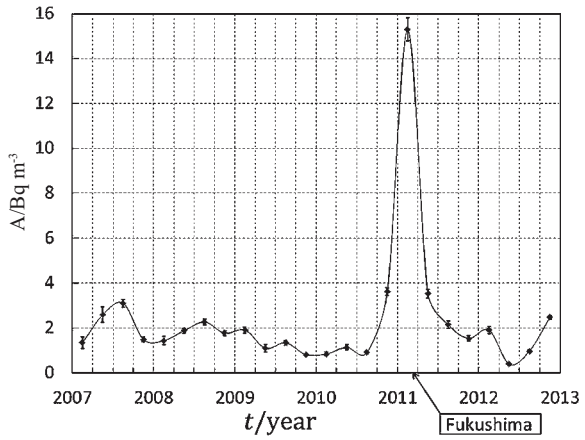

Figure 1 *Activity concentrations of ^{137}Cs in fallout.*

In contrast to the results for soil, where the increase of A over $\overline{A}_{\text{soil},07-10}$ was moderate, A in the fallout exhibited a pronounced peak in the spring of 2011. If we calculate the mean value of A before the accident using weighted statistics for all of the data prior to the peak, the result will amount to (0.875±0.009) Bq m^{-3}, which is about 16.5 times less than A_{max}. After the accident, A returned relatively quickly to the same level as before the accident.

The increase of A due to the accident was obviously much stronger in the fallout than in the soil. On the other hand, the effects of the pollution remained visible in the soil throughout 2012, but disappeared in the fallout relatively quickly following the accident. This implies that one can distinguish between short-term threats (from fallout) and long-term threats (from soil) to human health regarding pollution with ^{137}Cs originating from food.

Fortunately, in the case of the ^{137}Cs that reached Croatia from Fukushima, its amount in soil and fallout was not at a level that should raise alarm about excessive radiotoxic loads to the human body. During the transfer of ^{137}Cs from soil and fallout to food, its concentration decreased enough (5-10) to keep activity concentrations well below the allowed upper limit for food (600 Bq kg^{-1}) (11). Nevertheless, to achieve the highest amount of certainty, further investigations are necessary.

CONCLUSIONS

The results of measurements performed in the period between January 2007 and December 2012 have shown that the activity concentrations of ^{137}Cs in Zagreb, Croatia peaked both in fallout and in soil shortly after the Fukushima accident occurred. This demonstrated that radioactive contamination resulting from nuclear accidents may spread over large distances. For soil, this effect was stronger in layers closer to the surface. The maximum increase was by 27 % in the topmost layer (0 cm to 5 cm below surface), 60 % in the intermediate layer (5 cm to 10 cm below surface), and 18 % in the deepest layer (10 cm to 15 cm below surface). As for fallout, the activity concentration exhibited a steep and strong (16.5 times) increase after the accident, and a relatively fast return to average values. This was in contrast to soil, where the activity concentration remained slightly enhanced in the upper two layers. Therefore, the two media responded differently to contamination by anthropogenic radionuclides, which indicates that future studies should adopt a truly comprehensive approach. In this paper, the effects on food consumed by humans were far too weak to cause any public health problems stemming from the radiotoxicity of ^{137}Cs.

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Sažetak

^{137}Cs U TLU I OBORINI OKO ZAGREBA U VRIJEME NESREĆE U FUKUSHIMI

Tema ovog rada je primjetno povećanje koncentracije aktivnosti ^{137}Cs u tlu i oborini na zagrebačkom području u vrijeme nesreće u Fukushimi u proljeće 2011. Tema je iznimno važna za javno zdravstvo jer je ^{137}Cs radioaktivan i vrlo toksičan uslijed dugog vremena poluraspada i kemijske sličnosti s kalijem. Povećanje koncentracije u oborini bilo je puno jače nego u tlu, ali je u tlu bila dulje prisutan. Iako zamjetan u našim mjerenjima, višak ^{137}Cs nije se proširio kroz hranidbeni lanac u količinama koje bi dovele do premašivanja dopuštenih granica radioaktivne kontaminacije u namirnicama. Međutim, za preciznije zaključke o aktivnosti ^{137}Cs potrebna su temeljitija i dugotrajnija mjerenja.

KLJUČNE RIJEČI: *gamaspektrometrija, javno zdravstvo, radioaktivna kontaminacija*

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