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Dietary items as possible sources of ^{137}Cs in large carnivores in the Gorski Kotar forest ecosystem, Western Croatia

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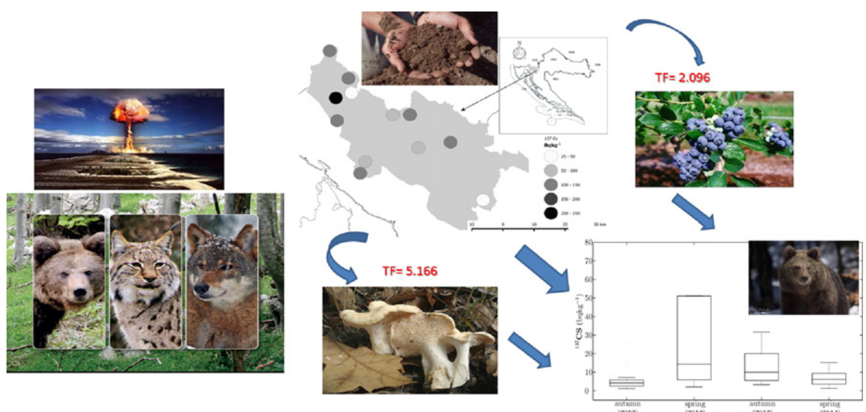
HIGHLIGHTS

- Radionuclide mass activities were determined by the gamma-spectrometric method.
- The highest ^{137}Cs mass activity in brown bear was 132, wolf 22.2 and lynx 153 Bq kg^{-1}.
- The best bioindicators are a wood hedgehog (TF = 5.166) and blueberry (TF = 2.096).

GRAPHICAL ABSTRACT



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ABSTRACT

The mountain forest ecosystem of Gorski Kotar is distant from any significant sources of environmental pollution, though recent findings have revealed that this region is among the most intense ^{137}Cs contaminated area in Croatia. Therefore, the aim of this study was to investigate ^{137}Cs and ^{40}K load in three large predator species in the mountain forest ecosystem. Radionuclides mass activities were determined by the gamma-spectrometric method in the muscle tissue of brown bear (47), wolf (7), lynx (1) and golden jackal (2). The highest ^{137}Cs mass activity was found in lynx (153 Bq kg^{-1}), followed by brown bear (132 Bq kg^{-1}), wolf (22.2 Bq kg^{-1}), and golden jackal (2.48 Bq kg^{-1}). Analysis of 63 samples of dietary items suggests that they are not all potentially dominant sources of ^{137}Cs for wildlife. The most important source of radionuclides for the higher parts of the food-chain from the study area were found to be the mushroom species wood hedgehog (*Hydnum repandum*), with a transfer factor TF of 5.166, and blueberry (*Vaccinium myrtillus*) as a plant species (TF = 2.096). Food items of animal origin indicated higher mass activity of radionuclides and therefore are possible moderate bioindicators of environmental pollution. The results also revealed that possible unknown wild animal food sources are a caesium source in the study region, and further study is required to illuminate this issue.

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1. Introduction

The bioaccumulation of radionuclides, above all ^{137}Cs, has been recorded throughout Europe following the Chernobyl accident in 1986. This bioaccumulation is the main anthropogenic environmental contaminant threat and can pose a public health problem (Barišić et al. 2002). Different countries, regions and ecosystems have received varying amounts of radionuclide contamination. Forest ecosystems suffered the most contamination and must therefore be given specific consideration (Calmon et al. 2009). Forests are very complex natural ecosystems where radionuclides persist longer among plants, mushroom and animals (Vilic et al. 2005; Semizhon et al. 2009; Vinichuk et al. 2010). Artificial radionuclide ^{137}Cs is the member of the same homologous series to which the ^{40}K belongs and, although ^{40}K is a naturally occurring radionuclide, the competitive effects of caesium and potassium cannot be excluded (Shaw and Bell 1991). The ^{137}Cs concentrations in different parts of the food chain primarily depend on the contamination level of the area, though some species can accumulate caesium in higher concentrations and as such can serve as good bioindicators. Mass activities (concentrations) of ^{137}Cs in many mushrooms have been found to be 10 to 100 times higher than in plants (Yoshida and Muramatsu 1998). Several authors have stated that mushrooms are among the dominant sources of ^{137}Cs for game animals (Hohmann and Huckschlag 2005; Steiner and Fielitz 2009; Kapala et al. 2015). Mushrooms have an enormous internal uptake area, mycelia, which are able to reach caesium not available to other plants, representing an important pathway by which ^{137}Cs enters the human food system (Vinichuk et al. 2010). Other forest fruits, especially berries that ripen throughout the year, should also be considered as a main source of high caesium mass activity (Zibold et al. 2001; Calmon et al. 2009). Moderate caesium mass activities have been recorded in earthworms, different roots and aboveground plant parts (leaves, twigs and needles), which also regular food items for forest wildlife (Lovrencic et al. 2008; Dvořák et al. 2010; Todorović et al. 2013). Different wild animals at the top of the food chain could be also considered as useful bioindicators of radionuclide environmental contamination, especially in forest ecosystems (Vilic et al. 2005). Resorption of caesium in the digestive tract of mammals is relatively high, and can reach up to 80% in ruminants and herbivores, or 100% in monogastric carnivores (Dvořák et al. 2010). Therefore, continuous monitoring of the mass activity of radionuclide isotopes in wild animals is important (Rispoli et al. 2014) in the evaluation of human exposure from consumption of contaminated meat (Šprem et al. 2013).

The mountain forest ecosystem of Gorski Kotar is relatively distant from any significant source of environmental pollution. The area is exposed mainly to pollutants deposited as fallout from global atmospheric contamination (Barišić et al. 2002). Recent findings reveal that this region of Croatia received the most intense radionuclide contamination from the release of radioactive isotopes in the past (Vilic et al. 2005; Lovrencic et al. 2008; Šprem et al. 2013).

The objectives of this study were: (*i*) to analyze the mass activity of ^{137}Cs and ^{40}K in the muscle tissue of large carnivores, and to examine whether age, gender, body mass or season had any influence on their appearance, primarily in brown bear (*Ursus arctos*), (*ii*) to explore the presence of ^{137}Cs (and ^{40}K — element homologous to caesium) in different food items from the forest ecosystem, and (*iii*) to examine which particular food items in the diet of large carnivores could be a potential bioindicator of caesium pollution.

2. Material and methods

2.1. Study area

The Gorski Kotar area is a mountainous region in western Croatia with a total surface area of roughly 1300 km^2 (Fig. 1). Small plateaus and fields, covered with brown or terra rossa soils developed on limestone and dolomite bedrock dominate the area. In general, these soils are characterized by high acidity, humus and potassium content, and poor phosphorus content. The region is covered by coniferous and beech forests (41%), composed mainly of silver fir (*Abies alba*), spruce (*Picea abies*) and beech (*Fagus sylvatica*), at elevations between 200 and 1534 m. The climate is mountainous with a Mediterranean influence: a mean annual temperature of 7.7 °C, a mean annual rainfall of 2079 mm and mean of 75 snow days (days with >5 cm of snow). The Gorski Kotar region is inhabited by tree large carnivore species: brown bear, wolf (*Canis lupus*) and Eurasian lynx (*Lynx lynx*).

2.2. Sampling, preparation, measurements and statistics

Carcasses of 47 brown bears [autumn season 2012; N = 14 (9♀; 5♂), spring season 2013; N = 13 (3♀; 10♂), autumn season 2013; N = 7 (2♀; 5♂), spring season 2014; N = 13 (6♀; 7♂)], seven wolves (5♀; 2♂), one female lynx and two male golden jackals (*Canis aureus*) were collected. These animals were either hunted during two hunting seasons (2012/2013 and 2013/2014), or perished in vehicle collisions or naturally in Gorski Kotar. Animal age was estimated using patterns of tooth replacement and eruption, and based on counting dentine layers on longitudinally sanded canine roots (Lombaard 1971; Rouichová and Andéra 2007). Body mass was measured for all individuals.

Additionally, samples of different food items (N = 63) were collected randomly around the study area based on the dietary habits of the studied species (Cicnjak et al. 1987; Stošić 1999; Krofel et al. 2011).

At least 500 g of tissue samples were taken from each individual animal for radionuclide analysis. Samples were cleaned of fat and connective tissues, placed in counting vessels (125 cm^3) of known geometry and kept frozen at a temperature of −18 °C until gamma-spectrometric analysis was conducted. All measured radionuclides mass activities of animal tissue (fresh weight) were recalculated on the day the animals were culled. Different samples of potential food items (e.g. fruits, chestnut, oak-tree, bogue-tree, dog rose, plum-tree, wild cherry, strawberries, honey, mushrooms, potatoes, ants, and worms) were collected during the years 2012, 2013 and 2014. Samples were fragmented and homogenized before being placed into counting vessels (125 cm^3) of known geometry, weighted and kept frozen at a temperature of −18 °C until gamma-spectrometric analysis was conducted.

Seven honeydew honey samples (from fir and spruce forests) were collected across the study area, due to frequent damage to beehives by brown bear (Sindičić et al. 2011). Samples of mixed (nectar and honeydew honey) and honeydew honey were mechanically collected during summers 2012 and 2013, by extracting honey from combs placed in the Gorski Kotar area. Honeydew honey types were identified by pollen analyses (Louveaux et al. 1978) and electrical conductivity measurements (Vorwohl 1964) with a multirange conductivity meter HI 8733 (Hanna Instruments). When pollen grains were present and the electrical conductivity was in the range 0.7–1.0 m Scm^{-1}, the sample was classified as mixed nectar and honeydew honey. When pollen grains were absent or very rare and electrical conductivity exceeded 1.0 mS cm^{-1}, the sample was classified as honeydew honey. After determination of honey type, the samples were placed in counting vessels of volume 125 cm^3 and known geometry, weighed and measured by the gamma-spectrometric method.

Finally, soil samples were randomly collected from 12 locations to a depth of 10 cm of open vertical soil profile. Soil samples were collected in 2007, 2010 and 2013. Soil content was homogenized, dried at 105 °C to a constant weight, the sieved fraction <2 mm was placed into counting vessels (125 cm^3) of known geometry, weighted and stored until gamma-spectrometric analysis was conducted.

All ^{137}Cs and ^{40}K mass activities measured in samples of different food items (fresh weight), honey and soils (dry weight) were recalculated on 1 July 2013.

Mass activities of ^{137}Cs and ^{40}K were determined by the gamma-spectrometric method using an HPGe semiconductor detector system