Tagami, Keiko; Howard, Brenda J.; Uchida, Shigeo. 2016.**The time-dependent transfer factor of radiocesium from soil to game animals in Japan after the Fukushima Dai-ichi nuclear accident**. *Environmental Science & Technology*, 50 (17). 9424-9431. 10.1021/acs.est.6b03011

The time-dependent transfer factor of radiocaesium from soil to game animals in Japan after the Fukushima Dai-ichi nuclear accident

Keiko Tagami[*1], Brenda J. Howard[2], Shigeo Uchida[1]*

[1]Office of Biospheric Assessment for Waste Disposal, National Institute of Radiological Sciences, National Institutes for Quantum and Radiological Science and Technology: Anagawa 4-9-1, Inage-ku, Chiba 263-8555, Japan

[2]Centre for Ecology and Hydrology, Lancaster Environment Centre: Library Avenue, Bailrigg, Lancaster, LA1 4AP, United Kingdom

*Corresponding author phone: +81 43 206 3256; fax: +81 43 206 3267; e-mail: tagami.keiko@qst.go.jp

KEYWORDS: Aggregated transfer factor; Caesium-137; Effective half-life; Wild boar; Sika deer; Asian black bear

ABSTRACT

Since the Fukushima Dai-ichi accident, monitoring of tissues from hunted game animals ensures compliance with the standard food limits for radionuclides in Japan. We quantified the transfer of ^{137}Cs from contaminated land to game animals using the Aggregated transfer factor (T_{ag} = activity concentration in meat [Bq kg^{-1} fw] / amount in soil [Bq m^{-2}]) of ^{137}Cs for Asian black bear, wild boar, sika deer, green pheasant, copper pheasant and wild duck, collected between 2011-2015. Open data sources were used from Fukushima, Miyagi, Ibaraki, Tochigi and Gunma prefectures. Our initially compiled data showed that the maximum reported ^{137}Cs activity concentration in wild boar after the Fukushima Dai-ichi accident were lower than those reported after the Chernobyl accident. The geometric mean T_{ag} values (m^{2}kg^{-1} fw) of ^{137}Cs in 2015 for Asian black bear, wild boar, sika deer and copper pheasant were similar $(1.9–5.1)\times10^{-3}$ while those for green pheasant and wild duck were about one order of magnitude lower at $(1.0–2.2)\times10^{-4}$. Effective half-lives were 1.2-6.9 y except for sika deer and copper pheasant where no decreases were found. In contrast to the Chernobyl accident, no seasonal change occurred in the meat ^{137}Cs activity concentrations of the wild animals during the study period.

INTRODUCTION

More than 5 years has passed since the Fukushima Dai-ichi Nuclear Power Plant (hereafter Fukushima Dai-ichi) accident occurred on March 11, 2011. In Fukushima Prefecture, forests cover 71% of the land area; many of these forests have been contaminated after the Fukushima Dai-ichi accident to varying extents.[1] Currently, the sale of wild food from forests, such as mushrooms, game animal meat, berries and edible new shoots of some trees, has been banned from areas which were highly contaminated by radiocaesium fallout,[2] because these foods often exceeded the standard limit for total radiocaesium (^{134}Cs and ^{137}Cs) of 100 Bq kg^{-1} fresh weight (fw).

Decontamination of the land in Fukushima and surrounding areas were initially carried out in the areas where people live and on agricultural land. The edge of forest areas was also decontaminated if they were close to houses or agricultural areas. However, large forest areas remained untreated[3] because they are less accessible to residents so decontamination of these areas would be less cost effective in decreasing external radiation to people and it would also have negative environmental side effects. In Japan, collection of wild foods from forests and other areas was an important recreational, as well as economically effective, activity for residents who lived near the contaminated forests, although the consumption amount was small.[4] In addition to the comprehensive food monitoring that is used, local food monitoring has been a practical and effective way to check the radioactive content of wild food and ensure that people avoid eating contaminated wild food exceeding food standard limits (> 100 Bq kg^{-1} fw) from the forests affected by the fallout from Fukushima Dai-ichi accident.[5]

To be able to inform local residents of areas where more contaminated wild food exceeding the food standard limit (> 100 Bq kg^{-1} fw) may occur, and to guide authorities in the management of the forest, it is important to quantify the transfer of radiocaesium from contaminated forests to wild food. The Aggregated transfer factor (T_{ag}), which is defined as radionuclide activity concentration in food (Bq kg^{-1} fw – for ^{137}Cs given as [^{137}Cs] in this paper) divided by the radionuclide ground deposition in soil (Bq m^{-2}), is commonly used to quantify transfer in extensive ecosystems such as forests.[6,7] The T_{ag} provides an estimate of the radiocaesium activity concentration in wild food collected in the forest based on the ground deposition of radiocaesium onto the forest floor. It therefore enables direct comparisons to be made between species, with respect to temporal and spatial factors by taking the varying deposition rates into account. The T_{ag} can also be used to assess internal radiation dose to animals.

In European countries, many studies on the fate of ^{137}Cs in forests were carried out after the Chernobyl accident, including the derivation of Aggregated transfer factors for wild food products. These values were collated in the parameter handbook published by IAEA[8] in Technical Report Series No. 472 based on review results by Calmon et al.[9] published in IAEA-TECDOC-1616. After the publication of the handbook, many additional related publications on the fate of ^{137}Cs in forest ecosystems were published.[10-16] The data from Chernobyl fallout studies in forests showed that the [^{137}Cs] decreases with time in many different forest compartments such as tree leaves and understory plants. However, that in wild animals such as wild boar and roe deer, [^{137}Cs] remained high for a long time, with seasonal peaks depending on the animals diet.[17-20] After the Fukushima Dai-ichi accident, T_{ag} values for different components of trees were reported[21-24] which decreased with time, as expected, due to processes such as the decreasing importance of direct interception of radiocaesium and a reduction in the bioavailable fraction in soils.[25,26] For wild animals, no information has yet been reported in international literature on the time dependency of T_{ag} for wild animals since the Fukushima Dai-ichi accident.

In this study, we have compiled T_{ag} data for ^{137}Cs in game meat collected from forest and other non-agricultural, extensive areas in the period 2011 to 2015. We have quantified how the ^{137}Cs T_{ag} values change with time by calculating effective half-lives in the game animals. By comparing these values with previously observed data after the Chernobyl accident, we consider whether the Chernobyl data are applicable for Japan after the Fukushima Dai-ichi accident.

MATERIALS AND METHODS

Animal ^{137}Cs data sources. Cs-137 activity concentrations in game meat data for Asian black bear (*Ursus thibetanus*), wild boar (*Sus scrofa*), sika deer (*Cervus nippon*), green pheasant (*Phasianus versicolor*), copper pheasant (*Syrmaticus soemmerringii*) and wild duck (*Anas poecilorhynch* and *Anas platyrhynchos*) were reported in food monitoring data from more than 10 prefectures after the Fukushima Dai-ichi accident. Because we needed soil ^{137}Cs ground deposition data (Bq m^{-2} to calculate T_{ag}, the game meat [^{137}Cs] data were only collated from areas where adequate nearby soil measurements were available. A map of the five prefectures, Fukushima, Miyagi, Ibaraki, Tochigi and Gunma, used for data collection is shown in Fig. 1. The following data sources were used for this analysis.

- Food monitoring (Ministry of Health Labor and Welfare[2])
- Wild animals monitoring (Fukushima Prefecture[27])
- Wild boar monitoring (Tochigi Prefecture[28])

The data collection period was from May 8, 2011 to December 31, 2015.

For game animals collected in Fukushima Prefecture, the location of each animal collection site was specified according to a Fukushima hunter map.[29] The hunter map uses 31×31 cells covering Fukushima Prefecture; one cell area is about 5.5 km (wide) × 4.7 km (long). The location where the wild animals were obtained for the other four prefectures, where radiocaesium deposition is less spatially variable, was allocated to the nearest local government specified location (cities, towns and villages).

To calculate T_{ag}, we used ^{137}Cs ($T_{1/2}$=30.17 y) because it has a longer physical half-life than ^{134}Cs ($T_{1/2}$=2.06 y). If only total radiocaesium (i.e. [^{134}Cs] and [^{137}Cs]) data were provided in the monitoring data, the contribution of ^{134}Cs was subtracted from the total radiocaesium using an assumed [^{134}Cs]:[^{137}Cs] activity ratio of 1:1 on March 11, 2011. All [^{137}Cs] were decay corrected to the date of measurement. Typically, the interval between sampling and measurement varied from 1-30 day so the delay in measurement was negligible due to the long physical half-life of ^{137}Cs.

To obtain T_{ag} values for each species from 2011 to 2015, further data selection was necessary because some meat samples had [^{137}Cs] below the detection limit. For these samples, if there were soil ^{137}Cs data in corresponding areas, then T_{ag} were reported as not detected, if not then the T_{ag} was not reported. A summary of the data is given in Table 1. The total number of T_{ag} values obtained for animals was relatively high for the mammals at 506 for Asian black bear, 2263 for wild boar, 470 for sika deer, and lower for the birds at 86 for green pheasant, 50 for copper pheasant and 140 for wild duck.

Corresponding soil data source. Soil ground deposition, in Bq m^{-2}, was obtained from open source data from the Ministry of Education, Culture, Sports, Science and Technologies (MEXT).[1] The soil values for ^{137}Cs (Bq m^{-2}) from MEXT, were plotted on a map of Japan as of June 14, 2011, March 1, 2012, September 1, 2012, December 1, 2012, July 1, 2013, December 1, 2013, and December 1, 2014. All data were decay corrected to the sampling date. The first intensive monitoring data were focused largely on the Fukushima Prefecture contaminated area, then subsequent monitoring campaigns covered other areas including Miyagi, Ibaraki, Tochigi and Gunma prefectures. For the first and second monitoring campaigns were carried out by collecting five soil samples at one sampling point and the mean value was provided for the sampling point. The following five campaigns were conducted by using standard in-situ Ge spectrometry for ^{137}Cs and ^{134}Cs determination (MEXT).[30]

Each wild game animal sampling site in Fukushima prefecture was located in the hunter map and the corresponding area in the ^{137}Cs soil ground deposition map was identified. If there was no soil data in the corresponding hunter map cell, then the soil data next to that cell (from any direction) were applied for the Fukushima samples. Outside Fukushima prefecture, the sampling sites of the animal samples in the four selected prefectures were at least 50 km from the Fukushima Dai-ichi site. Therefore, we assumed that the ^{137}Cs ground deposition in each local government level area was uniform (city, town and village). Thus, soil ground deposition ^{137}Cs data collected anywhere in the specified animal collection city/town/village were applied from the soil survey data map.

For soil ^{137}Cs ground deposition determination in all prefectures, if a single value was found in the corresponding area (cell or local government size), then the single value was used. If there were two data, then an average value was used, and a geometric mean (GM) value was applied if more than three soil data were available.

Data analysis. The Aggregated transfer factor, T_{ag}, was calculated using the following equation.

T_{ag} (m^2 kg^{-1} fw) = ^{137}Cs in meat (Bq kg^{-1} fw) / ^{137}Cs in soil (Bq m^{-2}) (1)

We also calculated effective half-life (T_{eff}) of [^{137}Cs] to compare with previously reported values after the Chernobyl accident. Effective half-life takes into account ecological and physical half-lives. To calculate T_{eff} of ^{137}Cs, the change with time [^{137}Cs] in a population in a natural ecosystem is generally used. However, as the samples were collected from areas with different soil ^{137}Cs radioactivity concentrations they were not appropriate for this calculation. To normalize [^{137}Cs], we simply applied the calculated T_{ag} data to calculate T_{eff}, which is defined as

$T_{eff} = \ln 2/\lambda_{eff}$ (2)

where λ_{eff} is the ^{137}Cs loss rate in each animal species. λ_{eff} is obtained from the slope of the exponential decline in [^{137}Cs] in the meat over time as follows:

$$A_t = A_0 \exp(-\lambda_{eff}\, t) \qquad (3)$$

where A_t is T_{ag} of ^{137}Cs at time t and A_0 is the expected initial T_{ag}. Using the [^{137}Cs] data, a best fit exponential trend line for each species was computed using KaleidaGraph software (Synergy Software, version 4.1.4).

RESULTS AND DISCUSSION

^{137}Cs activity concentrations in soil and game animals. In some areas data on the soil ^{137}Cs ground deposition was only available for one or two of the five years of the study period so we initially considered whether the available data were adequately representative of the entire sampling period of 2011-2015. We examined the change in ^{137}Cs ground deposition in soil from June 2011 to December 2014 using data collected at 150 sampling locations which had been measured on seven occasions in Fukushima Prefecture. The major soil type was brown forest soil, which occurs widely in the forests of the five prefectures considered in this study, with andosol, grey lowland soil and brown lowland soil also present. Both the GM of the seven ^{137}Cs ground deposition data at each sampling location and the ratio between each sampling time to the GM value was calculated.

The results are shown in Fig. 2; there was no statistically significant reduction in ^{137}Cs ground deposition over the period considered. Earlier data showed that in Japanese agricultural fields, the GMs of effective half-lives of global fallout ^{137}Cs were 18.1 y for paddy fields and 14.7 y for upland fields.[31] Thus, a longer observation period would be needed to detect a decrease in the ^{137}Cs ground deposition in forest areas. Therefore, we could reasonably assume that the ^{137}Cs ground deposition was similar between 2011-2015 and T_{ag} values were, therefore, directly comparable.

We initially examined whether the [^{137}Cs] in meat corresponded to the ^{137}Cs activity on the ground or not. Thus [^{137}Cs] data in each animal observed in 2011-2015 were classified into three different ^{137}Cs ground deposition categories in soil of 1-10 kBq m^{-2} (low), 10 - 100 kBq m^{-2} (middle), and >100 kBq m^{-2} (high). The results shown in Figure 3 indicated that the ground deposition of ^{137}Cs affected the [^{137}Cs] in game meat. The [^{137}Cs] in meat in the highly contaminated area were significantly higher than that in the less contaminated area except for sika deer (because there were no data in the >100 kBq m^{-2} area) and wild duck by Student's t-test. Amongst the data in each soil ^{137}Cs activity concentration category, wild boar [^{137}Cs] were significantly the most contaminated game animal sampled in high and middle categories, and higher than those of bird species in the low contamination category.

Howard et al.[7] summarized T_{ag} data for wild game animals and also reported [^{137}Cs] in game meat mostly collected in European countries. For wild boar, a maximum of 17.6 kBq kg^{-1} fw was recorded where the ^{137}Cs ground deposition to soil was 50 kBq m^{-2}. When wild boar data in Japan in the corresponding middle band of soil ^{137}Cs at 10-100 kBq m^{-2} were compared, a maximum value of 24 kBq kg^{-1} fw was observed. If we consider a smaller range which is more similar to the Chernobyl value above of 40 to 60 kBq m^{-2}, then the highest [^{137}Cs] in wild boar meat was 8.1 kBq kg^{-1}fw with a GM of 107 Bq kg^{-1}fw. Thus, the limited initial comparison indicated that the maximum [^{137}Cs] in wild boar was lower than that observed after the Chernobyl accident.

Aggregated transfer factor (T_{ag}). The determination of the underlying data for the deposition to soil is a key component in the use the T_{ag} value. The spatial resolution of the data is limited and the animals considered have different sizes of home range from which they derive their food, which introduces an averaging effect, but unavoidably includes uncertainties. Detailed studies on such uncertainties are currently being considered under the IAEA MODARIA programme,[32] but currently the use of T_{ag}, which amalgamates a large number of underlying processes, is a reasonable currently available approach which is widely used for radionuclide contamination.We have summarized the data in each year showing the number of T_{ag} values (m^2 kg^{-1} fw) obtained, GM, geometric standard deviation, and the range in Table 2. The percentage of detected to the total samples measured (see Table 1) was relatively low in wild duck in Fukushima prefecture (52%) and in green pheasant in the four less contaminated prefectures (45%), However, that for other combinations was consistently higher than 85%; especially for mammals where it was more than 95%. Because fewer measurable data were available for the birds and "less than" data were not used some probably low T_{ag} values are not included, so the T_{ag} reported here will be over estimated. The T_{ag} data for mammals are less affected by this issue. The annual GMs from 2011 to 2015 decreased slightly for wild boar (from 6.8×10^{-3} to 3.1×10^{-3}), green pheasant (from 8.9×10^{-4} to 1.0×10^{-4}) and wild duck (from 8.7×10^{-4} to 2.2×10^{-4}). In contrast, no change was evident for Asian black bear (from 5.2×10^{-3} to 4.2×10^{-3}), sika deer (from 7.2×10^{-3} to 5.1×10^{-3}), and copper pheasant (from 2.5×10^{-3} to 1.9×10^{-3}).

Using the most recent T_{ag} data in 2015, the data variation (maximum/minimum) among game animal species was compared. Maximum / minimum T_{ag} ratios were within one to two orders of magnitude except for wild boar where it varied by about four orders of magnitude. When we classified the wild boar T_{ag} data into each of the five prefectures, Fukushima prefecture had four orders of magnitude difference whereas for the other four prefectures the T_{ag} values were within two orders of magnitude. Because the higher variation closer to the Fukushima Dai-ichi site may be due to greater heterogeneity in ^{137}Cs ground deposition within each cell of the hunter map, which might cause over -and under-estimation of the T_{ag} value. Conversely, for the other four prefectures, where the land was contaminated more uniformly, the variation of T_{ag} was smaller for wild boar.

Koivurova et al.[33] measured radiocaesium in reindeer and moose in Northern Finland after the Fukushima Dai-ichi accident and found a similar T_{ag} to that after the Chernobyl accident. Thus, if source and environmental conditions were similar, then T_{ag} could be the same after these two accidents. We compared the T_{ag} values in Japan with the internationally collated value by IAEA in TECDOC-1616.[8] Only wild boar was reported in the IAEA data table and were directly comparable with our data. The T_{ag} range of GM values given within 5 y after the Chernobyl accident is 4×10^{-3} to 6.7×10^{-2} so the data obtained for wild boar in Japan were in the same range as the collated data by IAEA. There are IAEA data for red deer (*Cervus elaphus*), which is the same family as sika deer (*Cervus nippon*), with T_{ag} values of 3×10^{-2} to 5×10^{-2} observed in 1986-1991[7] which are slightly higher than our T_{ag} values for sika deer. Similarly, the GM IAEA value of pheasant (*Phasianus colchicus*), which is in the same family as green pheasant, was 3.2×10^{-4}; thus the T_{ag} is similar to the value we observed. For wild duck, waterfowl are a comparable group and the value consistently decreased from 1986 to 1989, from 1.3×10^{-2} to 2.4×10^{-3} in TECDOC-1616[8]; the TECDOC-1616 data are one to two orders of magnitude higher than this study. However, the IAEA TECDOC-1616 data for waterfowl has a wide range and the Japanese data are within, but at the lower end of, the reported range across the world as summarized by IAEA.

Statistical analysis was carried out to compare T_{ag} values obtained in 2011 and 2015 for the six animal species (ANOVA test), and the results are shown in Fig. 4. In 2011, T_{ag} values for mammals were significantly higher than those for birds, however, by 2015, copper pheasant data were of the same order of magnitude as that of the mammals. Sprem et al.[34] reported that [^{137}Cs] in omnivorous species of brown bear (*Ursus arctos*) and wild boar, were higher than those of herbivorous species, namely roe deer (*Capreolus capreolus*), red deer (*Cervus elaphus*) and chamois (*Rupicapra rupicapra*). However, our T_{ag} data showed no substantial differences among Asian black bear, wild boar and sika deer. Asian black bear, wild boar and the three bird species are omnivorous species and plants form an important part of their diet, sika deer is an herbivorous species.[35,36]

Copper pheasant and green pheasant size, both male and female are similar, however, it is not clear why the T_{ag} values of copper pheasant were higher than other bird species because little information is available on their diet. Furthermore, the number of data was too small for statistical analysis of time trends so more data collection is necessary for further analysis.

T_{eff} for game animals. The T_{ag} values were plotted against the sampling date (days after March 11, 2011) in Figure 5. Except for wild boar and sika deer, samples were not collected evenly throughout the years, but effective half-lives of these animals can be calculated using equations (2) and (3) using data during the collection period from 2011-2015. The T_{eff} data are summarized in Table 3. Although no correlations were found between time and log(T_{ag}) of sika deer and copper pheasant ($p>0.05$), we estimated T_{eff} using fitting from equation (3).

Two of the three types of birds had a relatively short effective half-life of T_{ag}-^{137}Cs of 1.2-1.9 y, however, copper pheasant, had a longer T_{eff}, although the data used for the estimation of T_{eff} were relatively low. Copper pheasants are normally located in mountain forests, while the wild duck live in freshwater and estuarine and green pheasant are usually located in grassy areas.[35, 37] In freshwater and estuarine water, [^{137}Cs] decreased rapidly after the Fukushima Dai-ichi accident[38] and thus ^{137}Cs sources to wild duck decreased faster than that in terrestrial areas. The difference may contribute to the relatively fast T_{eff} in wild duck compared with the other game animals. For green pheasant, the grassy areas that they inhabit were usually close to areas of human habitation which were decontaminated. These factors may have contributed in these two types of birds to the lower percentages of measureable [^{137}Cs] compared with the other four animals. Overall, there is lower confidence in the T_{eff} value for these birds and further data would be needed using longer counting times to obtain more reliable T_{eff} data for bird species.

The other animals had longer effective half-lives, especially sika deer for which no decrease was found. Pröhl et al.[39] reported T_{eff} of 10.5 y for wild boar (1986-1999) and 5.7-128 y for roe deer (1986-2002) after the Chernobyl accident. T_{eff} was also summarized in IAEA-TECDOC-1616[8] and the data obtained over a relatively early period after the Chernobyl accident (1986-1996) was 22 y, which is in agreement with the relatively high value we estimated for sika deer. Since our observation period was only for 5 years, and that there are a range of uncertainties relevant to the calculations made, a longer observation period would be necessary for a more informed and detailed data comparison with the data observed in European countries. However, it is clear that the [^{137}Cs] in Asian black bear, wild boar and sika deer has not declined much over the first few years after the accident.

Over three years after the Chernobyl accident, Karlén and Johanson[40] reported seasonal change of [^{137}Cs] in roe deer, which were higher in August to September than January to July. However, for sika deer, we did not find such a trend in the period 2011-2015. For wild boar meat, [^{137}Cs] data collected in 1988-1992,[19] and in 2001-2003[18] also showed a seasonal trend, which high values in summer and low in winter. However, as for sika deer, no seasonal trend was evident for wild boar in Japan. For both roe deer and wild boar in the Chernobyl fallout studies feeding on wild mushrooms and truffles (for wild boar) was the most significant contributor to high autumnal seasonal [^{137}Cs] in meat.[18,19,41] Consistent with the Chernobyl accident, in Japan the T_{ag} of [^{137}Cs] in mushrooms[24] showed the same order of magnitude as that reported in IAEA TRS 472,[9] and relatively high values have been reported in food monitoring data compared with agricultural food.[2] Therefore, if sika deer and wild boar in Japan ate mushrooms then their T_{ag} of ^{137}Cs should have increased in the fruiting period of mushrooms in these areas, i.e. September to November each year, but no such trend occurred. One possible reason for the lack of seasonal trends may be that mushrooms do not form an important part of the autumn diet for wild boar and sika deer in Japan contrasting with European animals. Available information supports this suggestion. Mushroom consumption was not recorded in wild boar in Japan by Asahi[42] or Kodera et al.[43] Also, only a trace amount of one fungal species was found in sika deer diet collected in June to August in Tochigi Prefecture,[44] and most of their diet consisted of tree parts and herbs.[36] To understand the fate of radiocaesium in game animals for a longer period of time, detailed research, such as that described by Hinton et al.,[45] of the ranging behavior and dietary habits of these game animals and the long-term behavior of radiocaesium in forest ecosystems will be needed in Fukushima Prefecture and surrounding areas.

ACKNOWLEDGMENTS

This work was partially supported by the Agency for Natural Resources and Energy, the Ministry of Economy, Trade and Industry (METI), Japan. It also formed part of the IAEA MODARIA programme WG4 on radioecological data.

REFERNCES

(1) Ministry of Education, Culture, Sports, Science and Technology Website; Extension Site of Distribution Map of Radiation Dose; http://ramap.jmc.or.jp/map/eng/map.html

(2) Ministry of Health, Labor and Welfare Website; Levels of Radioactive Contaminants in Foods Tested in Respective Prefectures; http://www.mhlw.go.jp/english/topics/2011eq/index_food_press.html

(3) Ministry of the Environment Website; Forest Environmental Remediation; http://josen.env.go.jp/about/efforts/forest.html

(4) Ministry of Health, Labor and Welfare Website; National Health and Nutrition Survey; http://www.mhlw.go.jp/seisakunitsuite/bunya/kenkou_iryou/kenkou/kenkounippon21/en/eiyouchousa/

(5) Taira, Y.; Hayashida, N.; Orita, M.; Yamaguchi, H.; Ide, J.; Endo, Y.; Yamashita, S.; Takamura, N. Evaluation of environmental contamination and estimated exposure doses after residents return home in Kawauchi Village, Fukushima Prefecture. *Environ. Sci. Technol*. **2014**, *48*, 4556-4563.

(6) Howard, B. J.; Beresford, N.;A.; Hove, K. Transfer of radiocesium to ruminants in unimproved natural and semi-natural ecosystems and appropriate countermeasures. *Health Phys.* **1991**, *61*, 715-725.

(7) Howard, B. J.; Johanson, K. J.; Linsley, G.;S.; Hove, K.; Pröhl, G.; Horyna, J. Transfer of radionuclides by Terrestrial food products from semi-natural ecosystems. In *Second Report of the VAMP Terrestrial Working Group*; IAEA-TECDOC-857. IAEA: Vienna 1996; pp 49-79.

(8) Calmon, P.; Thiry, Y.; Zibold, G.; Rantavaara, A.; Fesenko, S.; Orlov, O. Radionuclide transfer in forest ecosystems. In *Quantification of Radionuclide Transfer in Terrestrial and Freshwater Environments for Radiological Assessments*; IAEA-TECDOC-1616, IAEA: Vienna 2009; pp 333-380.

(9) IAEA. *Handbook of Parameter Values for the Prediction of Radionuclide Transfer in Terrestrial and Freshwater Environments*; Technical Report Series No. 472; IAEA: Vienna, 2010.

(10) Vinichuk, M.; Taylor A. F. S.; Rosén, K.; Johanson, K. J. Accumulation of potassium, rubidium and caesium (^{133}Cs and ^{137}Cs) in various fractions of soil and fungi in a Swedish forest. *Sci. Total Environ*. **2010**, *408*, 2543-2548.

(11) Karadeniz, Ö.; Yaprak, G. Soil-to-mushroom transfer of ^{137}Cs, ^{40}K, alkali-alkaline earth element and heavy metal in forest sites of Izmir, Turkey. *J. Radioanal. Nucl. Chem.* **2011**, *288*, 261-270.

(12) Thorring, H.; Skuterud, L.; Steinnes, E. Distribution and turnover of ^{137}Cs in birch forest ecosystems: influence of precipitation chemistry. *J. Environ. Radioactiv*. **2012**, *110*, 69-77.

(13) Lehto, J.; Vaaramaa, K.; Leskinen, A.^{137}Cs, ^{239,240}Pu and ^{241}Am in boreal forest soil and their transfer into widl mushrooms and berries. *J. Environ. Radioactiv.* **2013**, *116*, 124-132.

(14) Sheppard, S. C. Transfer factors to Whitetail deer: comparison of stomach-content, plant-sample and soil-sample concentrations as the denominator. *J. Environ. Radioactiv.* **2013**, *126*, 434-437.

(15) Skrkal, J.; Rulik, P.; Fantinova, K.; Burianova, J.; Helebrant, J. Long-term ^{137}Cs activity monitoring of mushrooms in forest ecosystems of the Czech Republic. *Radat. Prot. Dosim*. **2013**, *157*, 579-584.

(16) Kapla, J.; Mnich, K.; Mnich, S.; Karpinska, M.; Bielawska, A. Time-dependence of ^{137}Cs activity concentration in wild game meat in Knyszyn Primeval Forest (Poland). *J. Environ. Radioactiv.* **2015**, *141*, 76-81.

(17) Zibold, G.; Drissner, J.; Kaminski, S.; Klemt, E.; Miller, R. Time-dependence of the radiocaesium contamination of roe deer: measurement and modelling. *J. Environ. Radioactiv.* **2001**, *55*, 5-27.

(18) Hohmann, U.; Huckschlag D. Investigations on the radiocaesium contamination of wild boar (*Sus scrofa*) meat in Rhineland-Palatinate: a stomach content analysis. *Eur. J. Wildl Res.* **2005**, *51*, 263-270.

(19) Strebl, F.; Tataruch, F. Time trends (1986-2003) of radiocesium transfer to roe deer and wild boar in two Austrian forest regions, *J. Environ. Radioactiv.* **2007**, *98*, 137-152.

(20) Fielitz, U.; Klemt, E.; Strebl, F.; Tataruch, F.; Zibold, G. Seasonality of ^{137}Cs in roe deer from Austria and Germany. *J. Environ. Radioactiv.* **2009**, *100*, 241-249.

(21) Ramzaev, V.; Barkovsky A.; Goncharova, Yu.; Gromov, A.; Kaduka, M.; Romanovich, I. Radiocesium fallout in the grasslands on Sakhalin, Kunashiri and Shikotan islands due to Fukushima accident: the radioactive contamination of soil and plants in 2011. *J. Environ. Radioactiv.* **2013**, *118*, 128-142.

(22) Ohashi, S.; Okada, N.; Tanaka, A.; Nakai, W.; Takano, S. Radial and vertical distributions of radiocesium in tree stems of *Pinus densiflora* and *Quercus serrate* 1.5 y after the Fukushima nuclear disaster. *J. Environ. Radioactiv*. **2014**, *134*, 54-60.

(23) Renaud, Ph.;, Gonze, M. Lessons from the Fukushima and Chernobyl concerning the ^{137}Cs contamination of orchard fresh fruits. *Radioprotection* **2014**, *49*, 169-175.

(24) Nakai, W.; Okada, N.; Ohashi, S.; Tanaka, A. Evaluation of ^{137}Cs accumulation by mushrooms and trees based on the aggregated transfer factor. *J. Radioanal. Nucl. Chem.* **2015**, *303*, 2379-2389.

(25) Tagami, K.; Uchida, S. Effective half-lives of ^{137}Cs from persimmon tree tissue parts in Japan after Fukushima Dai-ichi Nuclear Power Plant accident. *J. Environ. Radioactiv.* **2015**, *141*, 8-33.

(26) Tagami, K.; Uchida, S. Effective half-lives of ^{137}Cs in giant butterbur and field horsetail, and the distribution differences of potassium and ^{137}Cs in aboveground tissue parts. *J. Environ. Radioactiv*. **2015**, *141*, 138-145.

(27) Fukushima Prefecture Website; Monitoring of Wild Animals; http://www.pref.fukushima.lg.jp/site/portal/wildlife-radiationmonitoring1.html (in Japanese)

(28) Tochigi Prefecture Website; Wild boar monitoring; http://www.pref.tochigi.lg.jp/g02/houdou/h23nakagawainoshishi.html (in Japanese).

(29) Fukushima Prefecture; Fukushima hunter map; http://www.pref.fukushima.lg.jp/uploaded/attachment/137276.pdf (in Japanese)

(30) Ministry of Education, Culture, Sports, Science and Technology. In-situ measurement using a Ge detecting system. Standard radioactivity method series No. 33; http://www.kankyo-hoshano.go.jp/series/lib/No33.pdf (in Japanese)

(31) Komamura, M.; Tsumira, A.; Yamaguchi, N.; Fujiwara, H.; Kihou, N.; Kodaira, K. Long-term monitoring and analysis of ^{90}Sr and ^{137}Cs concentrations in rice wheat and soils in Japan from 1959-2000. Technical Report No. 24 of National Institute for Agro-Environmental Sciences: Tsukuba, 2006; http://agriknowledge.affrc.go.jp/RN/2010724549.pdf (in Japanese).

(32) IAEA MODARIA Programme Website;

http://www-ns.iaea.org/projects/modaria/default.asp?l=116

(33) Koivurova, M.; Leppänen, A-P.; Kallio, A. Transfer factors and effective half-lives of ^{134}Cs and ^{137}Cs in different environmental sample types obtained from Northern Finland: case of Fukushima accident. *J. Environ. Radioactiv.* **2015**, 146, 73-79.

(34) Sprem, N.; Babic I.; Barisic, D.; Barisic. D. Concentration of ^{137}Cs and ^{40}K in meat of omnivore and herbivore game species in mountain forest ecosystems of Gorski Kotar, Croatia. *J. Radioanal. Nucl. Chem.* **2013**, *298*, 513-517.

(35) Ogasawara, K. Winter habitats and food habits of the green and copper pheasants. *J. Yamashina Inst. Ornithol.* **1968**, *5*, 351-362 (in Japanese).

(36) Ohdachi, S. D., Ishibashi, Y., Iwasa, M. A., Fukui, D., Saitoh, T., Eds. *The Wild Mammals of Japan*, 2nd, ed.; Shoukado: Tokyo, 2015.

(37) Higuchi, H.; Murai, H.; Hanawa, S.; Hamaya, S. The relationship between habitat characteristics and the abundance of waterfowl. *Strix (Journal of Wild Bird Society of japan)* **1988**, *7*, 193-202. (in Japanese)

(38) Nuclear Regulation Authority, Japan Website 2016; Monitoring information of environmental radioactivity level; http://radioactivity.nsr.go.jp/en/list/309/list-1.html

(39) Pröhl, G.; Ehlken, S.; Fiedler, I.; Kirchner, G.; Klemt, E.; Zibold, G. Ecological half-lives of ^{90}Sr and ^{137}Cs in terrestrial and aquatic ecosystems. *J. Environ. Radioactiv*. **2006**, *91*, 41-72.

(40) Karlén, G.; Johanson, K. J. Seasonal variation in the activity concentrations of ^{137}Cs in Swedish Roe-deer and in their daily intake. *J. Environ. Radioactiv*. **1991**, *14*, 91-103.

(41) Avila, R.; Johanson, K. J.; Bergström, R. Model of the seasonal variations of fungi ingestion and ^{137}Cs activity concentrations in roe deer. *J. Environ. Radioactiv*. **1999**, *46*, 99-112.

(42) Asahi, M. Stomach contents of wild boars (*Sus scrofa leucomystax*) in winter. *J. Mammalogical Soc. Japan* **1975**, *6*, 115-120 (in Japanese).

(43) Kodera, Y.; Kanzaki, N.; Ishikawa, N.; Minagawa, A. Food habits of wild boar (*Sus scrofa*) inhabiting Iwami District, Shimane Prefecture, western Japan. *Honyurui Kagaku (Mammalian Science)* **2013**, *53*, 279-287 (in Japanese).

(44) Marutama, N.; Totake, Y.; Katai, N. Seasonal change of food habits of the sika deer in Omote-Nikko. *J. Mammalogical Soc. Japan* **1975**, *6*, 163-173 (in Japanese).

(45) Hinton, T. G.; Byrne, M. E.; Webster, S.; Beasley, J. C. Quantifying the spatial and temporal variation in dose from external exposure to radiation: a new tool for use on free-ranging wildlife. *J. Environ. Radioactiv*. **2015**, *145*, 58-65.

432

Table 1. Data used for analysis of [^{137}Cs] in wild animals in Japan.

Animal name	Fukushima Prefecture			Miyagi, Ibaraki, Tochigi and Gunma Prefectures			Total number of T_{ag} calculated
	Number measured	Number detected (%)	Number of T_{ag} calculated	Number measured	Number detected (%)	Number of T_{ag} calculated	
Asian black bear	268	268 (100%)	268	268	267 (99.6%)	238	506
Wild boar	972	972 (100%)	972	1680	1630 (97%)	1291	2263
Sika deer	29	29 (100%)	29	467	443 (95%)	441	470
Green pheasant	69	69 (100%)	69	38	17 (45%)	17	86
Copper pheasant	39	39 (100%)	39	13	11 (85%)	11	50
Wild duck	102	53 (52%)	47	57	57 (100%)	57	104

Table 2. Cs-137 Aggregated transfer factor T_{ag} (m^2 kg^{-1} fw) of six wild animal species collected in Fukushima, Miyagi, Ibaraki, Tochigi and Gunma prefectures.

Animal name	Year	Number of samples	T_{ag} (m^2 kg^{-1} fw)			
			GM	GSD	Min	Max
Asian black bear	2011	23	5.2×10^{-3}	2.3	1.6×10^{-3}	4.7×10^{-2}
(*Ursus thibetanus*)	2012	160	3.9×10^{-3}	2.5	3.2×10^{-4}	6.7×10^{-2}
	2013	73	4.4×10^{-3}	2.5	9.1×10^{-4}	4.2×10^{-2}
	2014	191	2.8×10^{-3}	2.6	3.4×10^{-4}	8.0×10^{-2}
	2015	59	4.2×10^{-3}	2.2	6.0×10^{-4}	1.9×10^{-2}
Wild boar	2011	163	6.8×10^{-3}	2.7	4.7×10^{-4}	5.4×10^{-1}
(*Sus scrofa*)	2012	453	4.4×10^{-3}	2.6	2.1×10^{-4}	1.2×10^{0}
	2013	499	4.3×10^{-3}	3.3	2.9×10^{-4}	1.5×10^{-1}
	2014	543	2.6×10^{-3}	2.6	2.4×10^{-4}	8.3×10^{-2}
	2015	605	3.1×10^{-3}	2.8	8.9×10^{-5}	2.9×10^{-1}
Sika deer	2011	35	7.2×10^{-3}	2.7	1.3×10^{-3}	2.3×10^{-1}
(*Cervus nippon*)	2012	102	5.6×10^{-3}	2.5	5.1×10^{-4}	1.2×10^{-1}
	2013	131	5.5×10^{-3}	2.5	9.5×10^{-4}	7.5×10^{-2}
	2014	104	6.3×10^{-3}	3.1	4.6×10^{-4}	8.4×10^{-2}
	2015	98	5.1×10^{-3}	2.9	8.4×10^{-4}	3.7×10^{-2}
Green pheasant	2011	27	8.9×10^{-4}	2.4	2.7×10^{-4}	7.7×10^{-3}
(*Phasianus versicolor*)	2012	37	8.1×10^{-4}	2.6	5.9×10^{-5}	3.8×10^{-3}
	2013	12	2.7×10^{-4}	3.2	3.3×10^{-5}	1.2×10^{-3}
	2014	6	3.3×10^{-4}	2.4	1.2×10^{-4}	8.6×10^{-4}

	2015	4	1.0×10^{-4}	2.6	5.4×10^{-5}	4.2×10^{-4}
Copper pheasant	2011	10	2.5×10^{-3}	2.2	7.3×10^{-4}	9.0×10^{-3}
(*Syrmaticus soemmerringii*)	2012	21	1.6×10^{-3}	2.8	2.5×10^{-4}	1.3×10^{-2}
	2013	8	4.8×10^{-3}	3.0	8.3×10^{-4}	3.4×10^{-2}
	2014	6	1.7×10^{-3}	3.4	6.1×10^{-4}	1.1×10^{-2}
	2015	5	1.9×10^{-3}	1.7	1.3×10^{-3}	4.6×10^{-3}
Wild duck, incl.	2011	16	8.7×10^{-4}	2.0	1.9×10^{-4}	2.9×10^{-3}
Gray duck (*Anas zonorhyncha)*	2012	46	6.6×10^{-4}	2.9	9.3×10^{-5}	2.3×10^{-2}
Mallard (*Anas platyrhynchos*)	2013	18	5.5×10^{-4}	2.5	1.0×10^{-4}	2.3×10^{-3}
	2014	14	2.7×10^{-4}	2.2	7.4×10^{-5}	8.7×10^{-4}
	2015	10	2.2×10^{-4}	2.3	5.3×10^{-5}	7.0×10^{-4}

Table 3. Correlations of log(T_{ag}) with time and effective half lives (T_{eff}) for six game animals in Japan in 2011-2015.

Animal name	R	p	A_0	λ_{eff}	T_{eff}, y
Asian black bear	-0.119	0.0074	0.0047	0.00027	6.9
Wild boar	-0.227	<0.0001	0.0062	0.00053	3.6
Sika deer	-	0.856	0.0056	0.00002	(101)*
Green pheasant	-0.561	<0.0001	0.0016	0.00165	1.2
Copper pheasant	-	0.674	0.0024	0.00015	(13.1)*
Wild duck	-0.405	<0.0001	0.0011	0.00098	1.9

* Rough estimation values because no significant correlation with time for log(T_{ag}).

Figure 1. Map showing the location of the prefectures where wild game animal data were collected in 2011-2015.

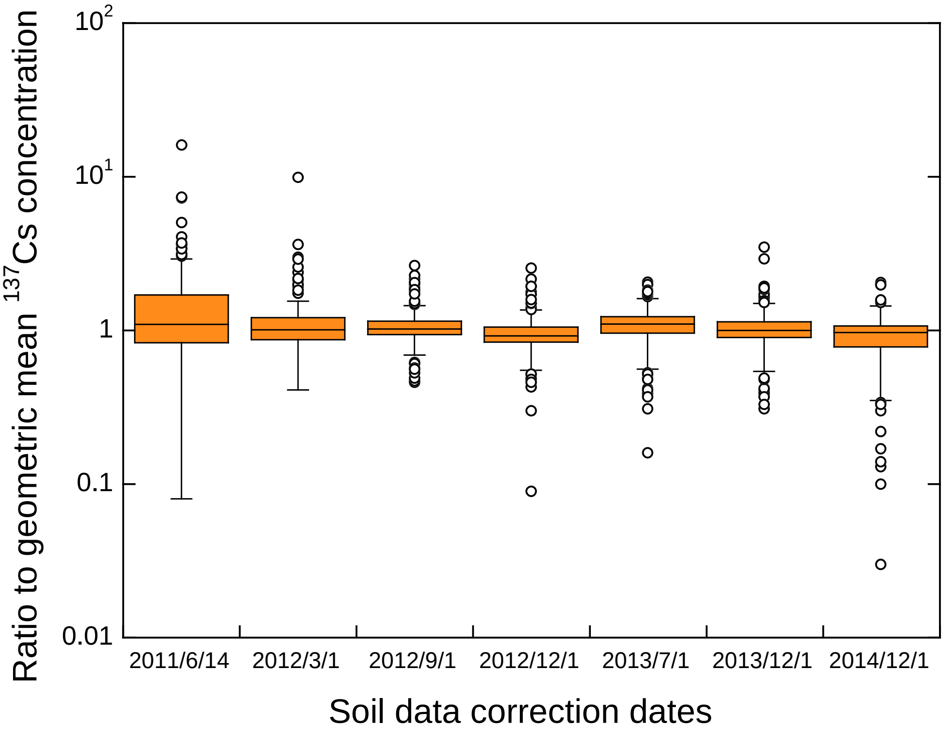

Figure 2. Ratio of the ^{137}Cs ground deposition at each sampling time to the geometric mean value at 150 sampling locations for seven sample occasions from 2011 to 2014.

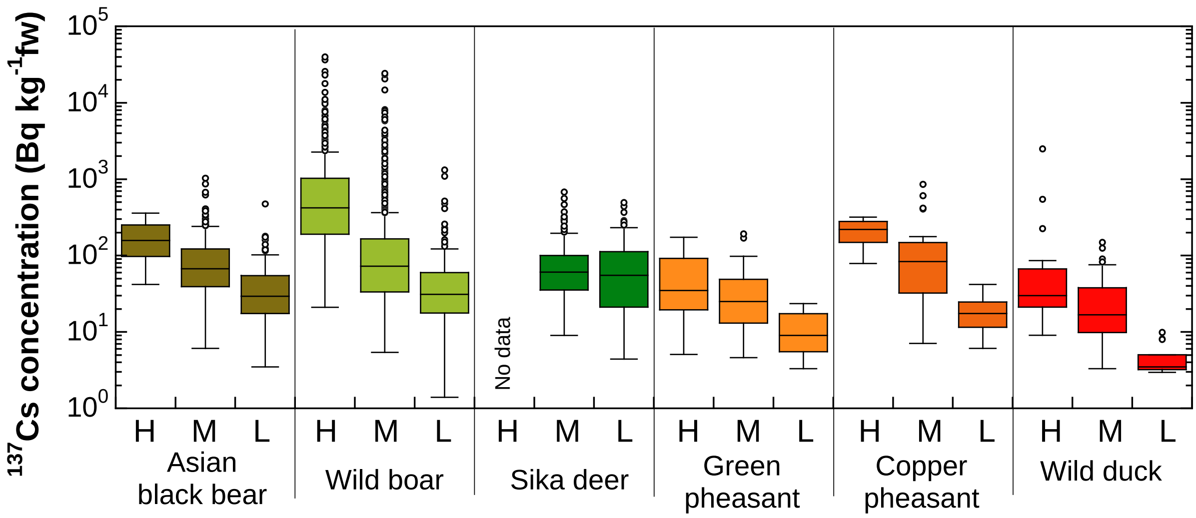

Figure 3. Cs-137 activity concentrations in meat of six game animals collected in high, medium and low bands of ^{137}Cs ground deposition in soil for the five most contaminated prefectures in Japan (H: >100 kBq m^{-2}, M: 10-100 kBq m^{-2}, L: 1-10 kBq m^{-2}).

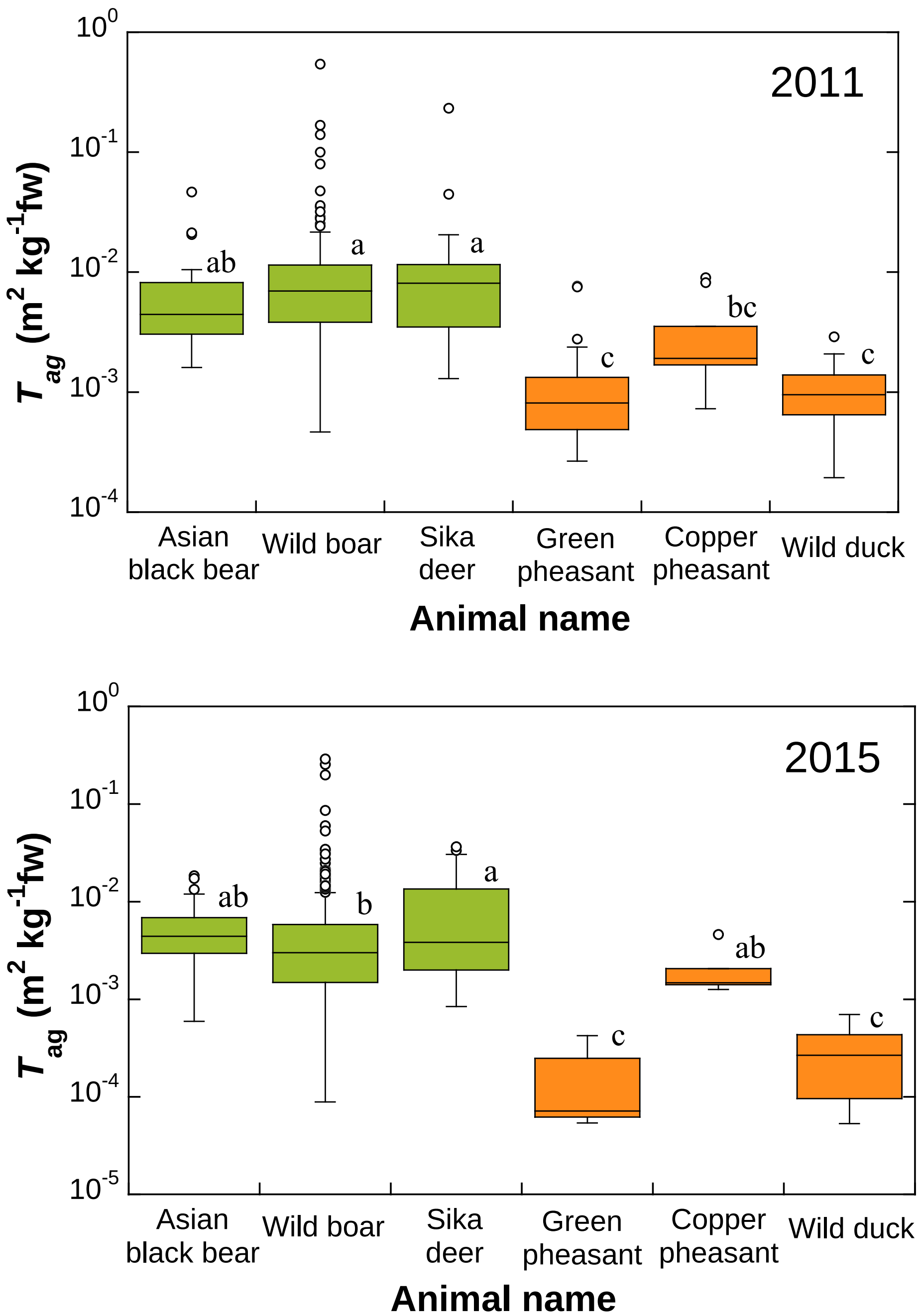

Figure 4. Comparison of T_{ag} (m^2 kg^{-1} fw) values in meat of six game animals collected in 2011 and 2015. Data with the same letter are not significantly different (p<0.05).

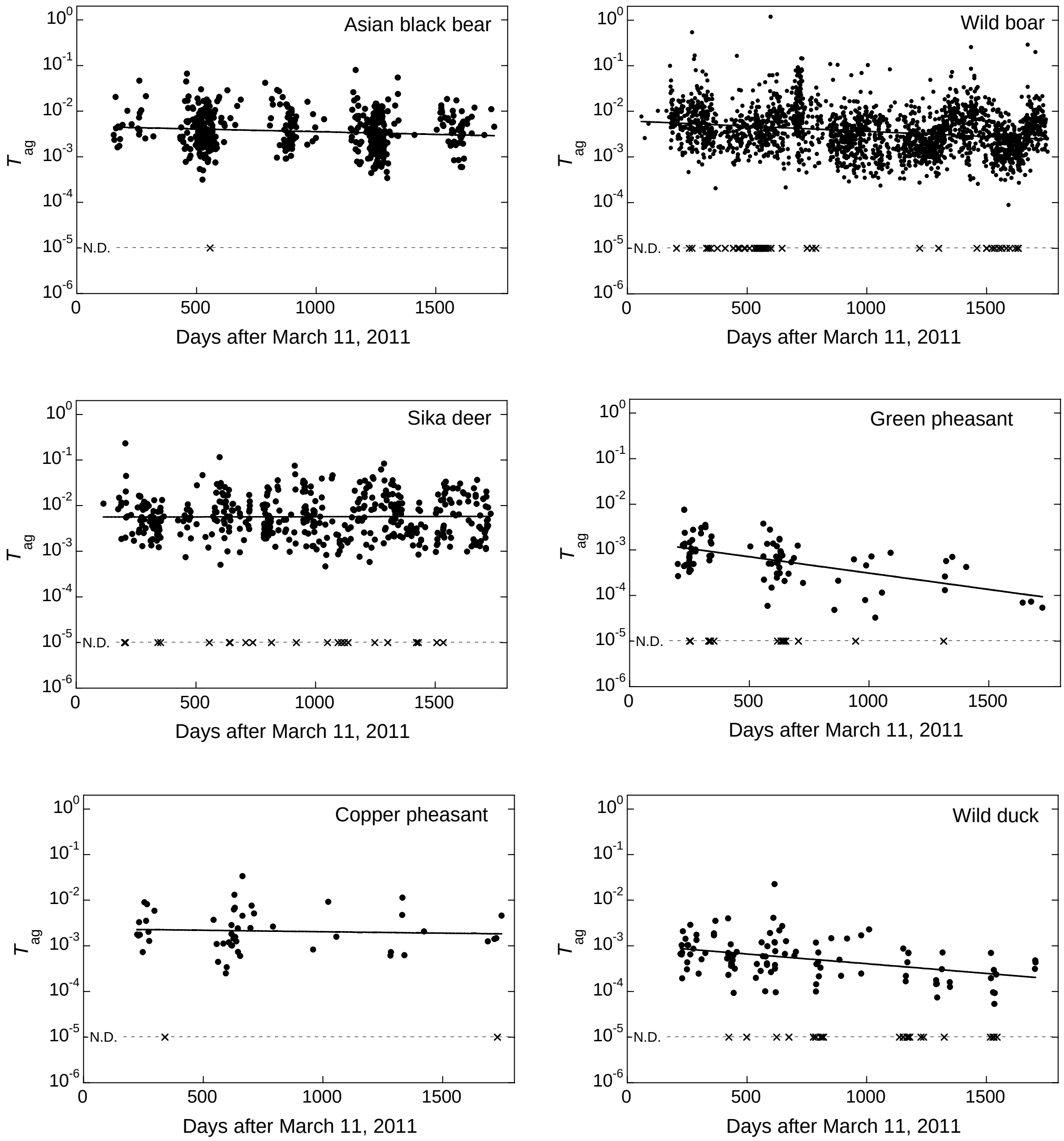

Figure 5. Time dependences of T_{ag} (m^2 kg^{-1} fw) of [^{137}Cs] in meat samples of Asian black bear, wild boar, sika deer, green pheasant, copper pheasant and wild duck. For the measurements with activity concentrations below the detection limit, a value of 10^{-5} was assumed.

Table of content/abstract graphic