Using Animal Thyroids as Ultra-Sensitive Biomonitorstrs for Environmental Radioiodine

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Supporting Information

ABSTRACT: In the course of the Fukushima nuclear accident large amounts of radionuclides relevant to the Comprehensive Nuclear-Test-Ban-Treaty (CTBT) were released and detected globally. We could show that the intake of environmental 131I into the thyroids of animals can be used for verification of the CTBT. Due to continuous accumulation of 131I, its apparent half-life in the thyroid biomonitor exceeds the physical one, thus making 131I detectable three weeks longer than using conventional CTBT-grade high volume air samplers. The maximum 131I activity concentrations (in Bq/kg) found in Austrian animal thyroids after the Fukushima nuclear accident could be correlated with the maximum activity concentrations found in air (Bq/m³) in Austria via a factor of 1.1 × 10⁶. In fall 2011, a second (much smaller) release of 131I occurred from a laboratory in Hungary, where this factor was 1.9 × 10⁶. Hence thyroid biomonitorstrs offer even some quantitative information, which allows the estimation of the 131I activity concentrations in air. It could be shown that thyroid biomonitorstrs can work under dry conditions, which potentially makes them the method of choice for CTBTO on-site inspections even in arid environments.

INTRODUCTION

The Comprehensive Nuclear-Test-Ban Treaty (CTBT) aims to ban nuclear test explosions in all environments. Once the treaty comes into effect, scientific verification of compliance with the treaty is of major importance (Article IV). Therefore, an International Monitoring System (IMS) is currently being established by the Preparatory Commission for the Comprehensive Nuclear-Test-Ban Treaty Organization (CTBTO) to create a worldwide monitoring network. It will consist of 321 monitoring stations and 16 radionuclide laboratories worldwide to monitor the underground, under water and the atmosphere for signs of a nuclear explosion. The IMS includes both geophysical (seismic, hydro-acoustic, infrasonic) and radioanalytical methods in order to impede clandestine nuclear weapon tests of at least 1 kt TNT equivalent without being detected by any of the stations anywhere on the globe with a probability of 90% within 10 days.1,2

Whereas the geophysical methods are used to detect and localize explosions, the monitoring of fission products released by nuclear weapon tests is crucial for the confirmation of the nuclear character of an explosion. Since nuclear activities do not necessarily yield a detectable seismic signal, as recently shown in Eastern Asia,3 sensitive radionuclide monitoring is becoming increasingly important. The IMS comprises technologies to monitor atmospheric fission and activation products in form of aerosols (140Ba, 131I and many others)4–8 as well as noble gases (37Ar and the xenon radioisotopes 131mXe, 133mXe, 133Xe, and 135Xe).9–11 The IMS is designed to detect traces of radionuclides even in great distances.12

Radioiodine (primarily 131I; half-life T1/2 = 8.03 days) is of great importance in the IMS as well for on-site inspections. After approximately 10% of the underground nuclear tests at the Nevada Test Site radionuclide leakage was observed, and in over 50% of those cases radioiodine was detected, with released quantities up to 10¹⁵ Bq.13 In the past, several kinds of environmental media were studied as monitors of radioactive fallout from nuclear weapon tests or nuclear accidents, including soil,14 fungi,15 lichens,16 tuna,17 canopy blades18 and many others.

Since the 1950s thyroids of animals have been studied as biomonitorstres for environmental radioactivity after nuclear tests and accidents.19–29 The pioneer in these studies was Lester Van Middlesworth (University of Tennessee), who dedicated five decades of his work to this topic. He investigated the intake of radioiodine into the thyroid with a countless number of...
individuals of several species of all ages and on several continents in order to gain fundamental knowledge on the accumulation of radioiodine in thyroids. His conclusions marked important contributions to the now established models of radiological protection. Most of his investigations were conducted after atmospheric nuclear tests, which naturally release greater amounts of fission products into the atmosphere than underground tests.

The Fukushima nuclear accident marked one of the rare examples of large amounts of radioiodine being released from a civil source into the environment. Consequently, CTBT-relevant 131I was monitored by (IMS-) stations all over the northern hemisphere. The objective of the present study was to investigate whether and how the process of 131I accumulation in animal thyroids meets the sensitivity criteria necessary for CTBT-verification and how animal thyroids can be used as an alternative and easily available low-tech monitor for the quantification of environmental radioiodine.

**EXPERIMENTAL SECTION**

**Animal Thyroids.** Thyroids were sampled from mainly grazing animals from eastern Austria in the periods March-June 2011 and October-November 2011. The animals were sacrificed for disease control purposes or were submitted to the pathology from zoos or private individuals. After dissection, the thyroids were measured immediately on a γ-spectrometer or deep frozen until measurement.

**Air Sampling.** Air was sampled in Vienna and Lower Austria. The particulate fraction of airborne radioiodine was filtered through conventional glass fiber filters using IMS-compatible high volume air samplers. For sampling of gaseous radioiodine in air, activated charcoal traps were used.

**Gamma Spectrometry.** Iodine-131 activity levels in thyroids were measured with a low-level γ-spectrometry facility, consisting of a 226 cm³ HPGe-detector (Canberra, detector model GC5020; 2.0 keV resolution at the 1332 keV 60Co peak; 52.8% relative efficiency), connected to a PC-based multi-channel analyzer with preloaded filter and located inside an ORTEC™ HBLBS1 shielding. Air samples were also measured on a low-level γ-spectrometer. For each sample geometry, the γ-detector’s efficiency was calibrated using a QCY48 (Amersham Ltd.) certified standard solution. The half-life of 131I is 8.03 days; it emits a unique γ-ray with an energy of 364.5 keV that is used for identification and quantification.

**RESULTS AND DISCUSSION**

During the Fukushima nuclear accident large quantities of CTBT-relevant nuclides were released, including 131I with source term estimations around some 10^{17} Bq. Approximately 10^{15} Bq of the released 131I passed over Europe, resulting in 131I activity concentrations in air in the mBq·m^{-3} range.

Sampling of animal thyroids started on 30 March 2011, approximately 10 days after contaminated air masses had
arrived over Europe. Of all animals investigated (see Supporting Information (SI Table S1), wild grazing animals (European moullon, Ovis orientalis musimon; red deer, Cervus elaphus; roe deer, Capreolus capreolus; European brown hare, Lepus europaeus; chamois, Rupicapra rupicapra) proved to be the best suited biomonitor. Others such as zoo animals, carnivores or omnivores (e.g., European badgers; Meles meles) did not or only in rare occasions show any radioiodine in detectable activity concentrations.

Results of the activity measurements in thyroids are shown in Figure 1. For tabulated results see SI Tables S1 and S2. For comparison, the activity levels in air at the corresponding dates (see SI Tables S3–S7) are included in Figure 1 as well.

The diagonal lines (indicating the physical decay behavior of $^{131}$I) in Figure 1 illustrate that the activity in a certain environmental medium is a function of more than one process. The combination of the physical radiological decay and loss processes of radionuclides in an ecosystem changes the effective decay characteristics of a radionuclide in a sample. This will lead to an effective half-life ($T_{\text{eff}}$) that will be shorter than the physical half-life. Iodine-131 activity concentrations in air exhibited a shorter effective half-life than the physical half-life of 8.03 days ($T_{\text{eff}} \sim 4–6$ days), which is due to dilution and wash-out effects (counterbalancing the arrival of fresh activity via atmospheric airflows). The same is true for the activity concentration observed in rainwater (Figure 1).

In animals, however, a third component accrues; namely active intake and accumulation in the thyroid. This, again, changes the apparent decay characteristics and leads to an apparent half-life that (at least for a certain time span) is longer than the effective or even the physical half-life. The apparent half-life ($T_{\text{app}}$) of $^{131}$I in thyroids proved to be as long as at least 9 days when calculated from the maximum activity (see SI Figure S1), which is due to continuous accumulation, counter-balancing the losses due to physical decay, atmospheric dilution, wash-out with rain and metabolic processes. This $T_{\text{app}}$ value compares well with the $\sim 10.5$ days reported for the $T_{\text{app}}$ of $^{131}$I in the thyroids of cattle after a nuclear test.21

After the Fukushima accident, the CTBTO’s monitoring station on the rooftop of the Vienna International Centre reported the latest detection of airborne $^{131}$I on 7 May 2011. The latest detection by an Austrian governmental monitoring station (Retz; northern Lower Austria) was reported on 16 May 2011. As a consequence of the extended apparent half-life, $^{131}$I remained detectable in thyroids until 6 June 2011, that is, at least three weeks longer than in air using CTBT-grade high volume samplers. After three weeks—that is, almost three half-lives of $^{131}$I—any arbitrary $^{131}$I activity will have dropped to 16% of its initial level only due to physical decay (not yet taking into account dilution effects, which will cause an even greater decrease of the effective activity). This impressively illustrates the high sensitivity of thyroid biomonitor compared with conventional systems.

Despite some fluctuations that are all roughly within one order or magnitude, it can be seen in Figure 1 that the time of sacrifice marks a more important feature for the resulting activity concentration in thyroids than the nature of the investigated species. One can assume that bigger animals that graze larger amounts of contaminated pasture also have bigger thyroid glands, which causes comparable activity concentrations in the thyroids of small and big grazing animals. A detailed discussion of the differences in the activity concentrations between different species is given elsewhere.22 Of course biological systems such as animal thyroids are always subject to certain fluctuations that are caused by the complexity of the animals’ metabolism.

In October/November 2011, an independent release of radioiodine into the atmosphere was reported by several European monitoring stations, which could finally be attributed to an accidental release from a radionuclide laboratory in Hungary.41 According to the Austrian Federal Ministry for Agriculture, Forestry, Environment and Water Management, a total of 370 GBq $^{131}$I were released in that case, leading to activity concentrations in eastern Austria in the range of tens of microbecquerel per m$^3$.

Although a mobile sampler (Sibata HVC-1000N; flow rate 60 m$^3$·h$^{-1}$) provided by the CTBTO was stationed on the roof of the Atominstitut in Vienna at the time of this event, the $^{131}$I activity levels did not exceed the detection limit. This was probably due to the limited flow rate, which did not meet the CTBTO criteria of 500 m$^3$·h$^{-1}$. However, several but not all CTBTO-grade European stations reported detections of radioiodine at that time. In any case, also the grazing mammals’ thyroids measured at that time revealed detectable $^{131}$I activity concentrations (Figure 1, SI Table S2).

Comparison of the correlation between the maximum $^{131}$I activity concentrations found in both air and thyroids shows decent agreement between both events, $^{131}$I from the Fukushima accident as well as from the Hungarian event. Comparing the maximum values was deemed the easiest way of correlating the two dynamic processes with unknown delay in-between. The obtained correlation factors between activity concentration in air (Bq·m$^{-3}$) and thyroids (Bq·kg$^{-1}$) was $1.1 \times 10^6$ in the course of the Fukushima nuclear accident, and $1.9 \times 10^6$ in the course of the Hungarian event. We define the correlation factors as the ratios of activity concentration in thyroid to that in air. One can use this correlation factor as a kind of “calibration” of the thyroid system for a rough estimation of the $^{131}$I activity level in air by measuring the respective activity concentration in animal thyroids.

It is important to note the fall 2011 was exceptionally dry with no rainfall in Austria during the entire time frame of the Hungarian event. Rainfall is known to scavenge (radioiodine) particles onto grassland. Wet deposition, therefore has always been regarded as the most important route of radioiodine fallout onto pasture. Our study, in contrast to previous expectations, confirms that the process of $^{131}$I accumulation in animal thyroids also occurs under such dry conditions. Even further, the above-mentioned “calibration” of the thyroid biomonitor for the estimation of the activity concentration in air from the activity concentration in a thyroid, which was obtained in the course of the Fukushima plume over Europe, was found to produce similar results under dry conditions as well. This implies that animal thyroid biomonitor can also work under dry conditions, which may be of importance for CTBTO-conducted on-site inspections in arid areas. Consequently, thyroid monitors are superior to monitoring precipitation (or other inanimate environments), because animals incorporate both wet as well as dry deposited radioiodine into their thyroids. This fact also makes them suitable for human consequence prediction after releases of $^{131}$I into the environment.

In January 2012 another $^{131}$I-release from the same Hungarian radionuclide laboratory was reported. These releases, however, were only approximately one tenth of the October/November 2011 event and could barely be monitored...
by any European monitoring stations. In January/February 2012, weather conditions in Austria were extremely frosty and snowy. In this case only one thyroid of a roe deer was provided for measurement, which revealed no detectable $^{131}$I levels. Together with the fact that the thyroids of carnivores and omnivores investigated during the Fukushima period did not contain detectable $^{131}$I concentrations, this observation allows the conclusion that respiratory intake of $^{131}$I—although it was shown to occur$^{39,42}$—is a minor path (at least at low activity concentrations) into the animal thyroid compared with the ingestion of contaminated pasture and water. A previous study$^{31}$ showed that $^{131}$I is effectively scavenged by soil systems, making it bioavailable to plants. Although much of the behavior of radioiodine in the environment is not yet fully understood, it appears likely that, given its short half-life, much of the $^{131}$I is surgically adsorbed on the leaves of the pasture and not actively taken up by the roots of the plants prior to animal intake. The lack of rainfall during the Hungarian event in 2011 also may be an indication that surficial adsorption of the fallout on plants can be a sufficient mechanism for the observed phenomena.

In both events, Fukushima and the Hungarian release, $^{131}$I was predominantly present in gaseous form (chemical species such as I$_2$ or organic halogenides).$^{33}$ Gaseous radioiodine is not washed out from the atmosphere by rainfall as efficiently as the particulate form.$^{36-38}$ However, the transfer of $^{131}$I from gaseous to particulate form—a process which takes weeks to occur—has been documented previously.$^{43}$ In other words, the factor time on the one hand increases the bioavailability of radioiodine by transfer from gaseous to particulate form, but, at the same time, it also decreases it due to physical decay and other loss processes on the other hand. In any case, the monitoring of gaseous $^{131}$I requires activated carbon traps, which usually have a much lower flow rate than conventional high volume samplers. Such traps are therefore not incorporated into the IMS. Thyroids proved to be highly sensitive biomonitors for atmospheric $^{131}$I despite its mainly gaseous form, indicating that the transfer from gaseous to particulate form is a sufficient process allowing accumulation.

The main sensitivity criterion currently applied to aerosol radionuclide stations of the IMS is a baseline sensitivity of 10 $\mu$Bq·m$^{-3}$ for $^{140}$Ba.$^4$ Required detection limits for airborne radioiodine are not regulated explicitly, but they are typically less than 1 order of magnitude lower (approximately 3 $\mu$Bq·m$^{-3}$).$^{44}$ With that activity concentration, one can estimate whether or not animal thyroid biomonitors will comply with the minimum detectable activity in the IMS. Assuming an activity concentration of 3 $\mu$Bq·m$^{-3}$ in air and using the correlation factor obtained in the course of the Fukushima accident, one can expect a $^{131}$I activity in the range of 6 mBq per (small) animal thyroid of 1.8 g. This absolute activity certainly can be detected using low-level $\gamma$-spectrometry, showing that animal thyroid biomonitors fulfill or even exceed the requirements for CTBT-verification.

The analysis for $^{131}$I of animal thyroids—preferably from (grazing) animals such as sheep, goats, cattle, pigs,$^{22}$ but also from wild herbivore animals such as mouflons (wild sheep) or deer—may be the only easily feasible opportunity to monitor the “smoking gun” of a nuclear explosion in the course of on-site inspections in remote areas that can only be accessed with a delay of weeks or months. Unexpectedly, thyroid biomonitors proved to also perform well under dry conditions, which makes them also potentially applicable in arid environments. The only restrictions of such monitors may be extreme climate such as cold and snowy weather conditions or other circumstances under which animals cannot graze.

## ASSOCIATED CONTENT

### Supporting Information

The estimation and illustration of the apparent half-life of $^{131}$I in animal thyroids is illustrated in Figure S1. The Tables S1 and S2 present tabulated $^{131}$I activity concentrations in animal thyroids after the Fukushima nuclear accident (S1) as well as after the Hungarian event (S2). The Tables S3–S7 present tabulated $^{131}$I activity concentrations in air in Austria: Alt-Prerau (particulate fraction; S3), Retz (particulate fraction; S4); Northern Vienna (particulate fraction; sampler A: S5 and sampler B: S6); Northern Vienna (gaseous fraction; S7). This material is available free of charge via the Internet at http://pubs.acs.org.

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### Notes

The authors declare no competing financial interest.

## ACKNOWLEDGMENTS

We thank Dr. Johannes H. Sterba for his support in the course of this study. Partial funding of this work by the Austrian Federal Ministry for Agriculture, Forestry, Environment and Water Management (BMLFUW) and the Dr. Michael-Häupl-Fonds is gratefully acknowledged.

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