Journal of Environmental Radioactivity 150 (2015) 20–35

Contents lists available at ScienceDirect

Journal of Environmental Radioactivity

journal homepage: www.elsevier.com/locate/jenvrad

Retrospective dosimetry of Iodine-131 exposures using Iodine-129 and Caesium-137 inventories in soils – A critical evaluation of the consequences of the Chernobyl accident in parts of Northern Ukraine

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ARTICLE INFO

Article history:
Received 26 April 2015
Received in revised form
20 July 2015
Accepted 22 July 2015
Available online 7 August 2015

Keywords:
Iodine-129
Iodine-131
Caesium-137
Accelerator mass spectrometry
ICP-MS
Retrospective dosimetry
Radionuclide migration

ABSTRACT

The radiation exposure of thyroid glands due to ^{131}I as a consequence of the Chernobyl accident was investigated retrospectively based on ^{129}I and ^{137}Cs inventories in soils in Northern Ukraine. To this end, soil samples from 60 settlements were investigated for ^{129}I, ^{127}I, and ^{137}Cs by AMS, ICP-MS and gamma-spectrometry, respectively. Sampling was performed between 2004 und 2007.

In those parts of Northern Ukraine investigated here the ^{129}I and ^{137}Cs inventories are well correlated, the variability of the individual ^{129}I/^{137}Cs ratios being, however, high. Both the ^{129}I and ^{137}Cs inventories in the individual 5 samples for each settlement allowed estimating the uncertainties of the inventories due to the variability of the radionuclide deposition and consequently of the retrospective dosimetry.

Thyroid equivalent doses were calculated from the ^{129}I and the ^{137}Cs inventories using aggregated dose coefficients for 5-year old and 10-year-old children as well as for adults. The highest thyroid equivalent doses (calculated from ^{129}I inventories) were calculated for Wladimirowka with 30 Gy for 5-years-old children and 7 Gy for adults. In 35 settlements of contamination zone II the geometric mean of the thyroid equivalent doses was 2.0 Gy for 5-years-old children with a geometric standard deviation (GSD) of 3.0. For adults the geometric mean was 0.47 Gy also with a GSD of 3.0. In more than 25 settlements of contamination zone III the geometric means were 0.82 Gy for 5-years old children with a GSD of 1.8 and 0.21 Gy for adults (GSD 1.8).

For 45 settlements, the results of the retrospective dosimetry could be compared with thyroid equivalent doses calculated using time-integrated ^{131}I activities of thyroids which were measured in 1986. Thus, a critical evaluation of the results was possible which demonstrated the general feasibility of the method, but also the associated uncertainties and limitations.

1. Introduction

This work is part of a radioecological project by which the environmental behaviour of various radionuclides released by the Chernobyl accident and the resulting radiation exposure of the inhabitants in the highly contaminated areas in Northern Ukraine were studied; e.g. Schmidt et al. (1998), Filss et al. (1998), Slavov et al. (1999), Botsch et al. (2000), Handl et al. (2003), Mewis (2004), and Hippler (2006). These investigations were aimed on the evaluation of the exposures of inhabitants of contamination zones II and III in Northern Ukraine due to ^{137}Cs, ^{90}Sr, and actinide isotopes more than a decade after the accident. With the goal of retrospective dosimetry of ^{131}I exposures shortly after the accident we analysed ^{127}I and ^{129}I in soils from Russia and Northern Ukraine (Michel et al., 2005). Results were obtained for exposure to ^{131}I of people living in the Korosten and Narodici districts (Zhitomir region) of Northern Ukraine.

After a reactor accident with massive releases of radionuclides to the environment, reliable estimates of the thyroid exposures due to inhalation and ingestion of ^{131}I are essential to assess the risk of

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http://dx.doi.org/10.1016/j.jenvrad.2015.07.023
0265-931X/

thyroid cancer. Repeated direct measurements of the ^{131}I activity in the thyroids of the affected population provide the best means for reliable estimates of the thyroid exposure. However, comprehensive measurements of the thyroid activities in an entire population are difficult to organize in the immediate aftermath of such an emergency. Moreover, the short half-life of ^{131}I (8.02 days) does not allow extending such measurements over larger time spans.

Consequently, one has to rely to a large degree on retrospective dosimetry on the basis of the environmental abundances of long-lived radionuclides released in the accident. After the Chernobyl accident, ^{129}I ($T_{1/2} = 15.7$ Ma) and ^{137}Cs ($T_{1/2} = 30.17$ a) were suitable radionuclides for this purpose. As volatile elements they were released in large amounts together with ^{131}I during the initial explosion, the subsequent graphite burning and melt-down of the residual core, and they were transported over large distances. By fall-out and wash-out large parts of the northern hemisphere and in particular of Europe were contaminated. Actinide elements and ^{90}Sr were not suited for this purpose since they were strongly fractionated relative to iodine in the release process due to their lower volatility. Also the transport, fall-out and wash-out processes of ^{90}Sr and actinide elements differ strongly from those of iodine and caesium. See UNSCEAR (2000) for a detailed assessment of the accident.

Undisturbed soils can be used as archives for the fall-out of long-lived radionuclides for the purpose of retrospective dosimetry. In order to retrospectively estimate radiation exposures using long-lived radionuclides in the soil archives, radioecological modelling is needed to describe the pathways of the radionuclides through the environment to humans. This appears reasonably feasible if the exposure is dominated by external irradiation or by the ingestion pathway since data on the actual behaviour (occupancy and shielding factors), consumption habits (e.g. milk and green vegetables) as well as land and animal use can be retrospectively surveyed. The contribution to the exposure via the inhalation pathway is more difficult to estimate since it requires information about the transition of the radioactive plumes and the actual weather conditions, which involves meteorological observations or modelling.

The analysis of the long-lived radionuclide ^{129}I in soils is – in principle – the best method for the retrospective dosimetry of the radiation exposure caused by ^{131}I long after the release in accidents or incidents because the physico-chemical properties of both nuclides are practically identical. However, there are several requirements for this retrospective dosimetry to be feasible. The ^{129}I from the accident must not have disappeared from the analysed soil horizons by migration into deeper soil layers and it must be distinguishable from potentially existing fall-out prior to accident. Finally, the radioecological modelling connecting the inventory of ^{131}I derived from the observed ^{129}I deposition densities with the thyroid dose must be as realistic as possible on the basis of the available information.

The environmental abundances of ^{129}I and its pathways through the different environmental compartments represent a scientifically quite complicated case because of its various natural and man-made sources and its poorly understood environmental chemistry. It is produced in nature by cosmic-ray induced spallation of xenon in the atmosphere and by spontaneous fission in the geosphere. The natural abundances of ^{129}I have been globally enhanced by ^{129}I released by man into the environment. Man-made ^{129}I originates primarily from induced fission of ^{235}U and ^{239}Pu. Explosions of nuclear weapons, the Chernobyl accident and, most importantly, releases from reprocessing plants have raised the environmental abundances locally and globally by orders of magnitude, although this radionuclide is not yet of radiological relevance except for the closest proximity to reprocessing plants. Man-made ^{129}I enters the environmental iodine cycles and changes the environmental iodine isotopic abundances. See Schmidt et al. (1998), Michel et al. (2005, 2012) for details and references.

There are several other publications dealing with the retrospective dosimetry of ^{131}I exposures using ^{129}I data. Straume et al. (1996, 2006), Robl et al. (1997), and Mironov et al. (1999, 2002) investigated the situation in Belarus, Pietrzak-Flis et al. (2003) in Poland. Endo et al. (2008) used ^{129}I for the retrospective dosimetry of the inhabitants of Dolon near Semipalatinsk in Kazakhstan. Dolon is close to the nuclear test site where 450 tests of nuclear weapons were performed between 1949 and 1989. Reithmeier et al. (2002) dealt with the general methodology of ^{131}I dose reconstruction in the former Soviet-Union by measurement of ^{129}I. Most of the earlier work aimed at evaluating the feasibility of ^{129}I retrospective dosimetry and on determining some of the input quantities needed. Only Robl et al. (1997) and Pietrzak-Flis et al. (2003) did complete evaluations ending up with thyroid dose estimates.

During recent years the retrospective dosimetry of ^{131}I exposures via ^{129}I has again become of high interest because the thyroid exposures after the Fukushima Dai-Ichi accident were insufficiently measured and have to be evaluated by retrospective dosimetry. Investigations with the goal of retrospective dosimetry are under way (Miyake et al., 2012).

More often than ^{129}I, ^{137}Cs deposition has been used to estimate thyroid exposures to ^{131}I after the Chernobyl accident because ^{137}Cs is much simpler to analyse. Kruk et al. (2004) summarized the respective earlier work and performed new model calculations for Belarus. They considered the actual weather and ecological conditions at the time of the accident and - as far as available - information about the fall-out ratio of ^{131}I and ^{137}Cs. The latter ratio is the crucial quantity if ^{137}Cs is used for the retrospective dosimetry of ^{131}I exposures. The radionuclides showed considerable variability due to time-dependent elemental fractionation during emission and atmospheric transport. UNSCEAR (2000) reported values between 10 and 30 for the $^{131}I/^{137}Cs$ ratio and Talerko (2005) calculated ratios of ^{131}I and ^{137}Cs inventories between 5 and 28 for Ukraine taking into account the detailed emission history and the actual weather conditions.

In this work, we continue our earlier investigations (Michel et al., 2005) by a systematic study of a large number of settlements in the contamination zones II and III of Northern Ukraine. The first goal was to obtain thyroid dose estimates for an area where excessive ^{131}I exposures occurred in 1986 combined with estimates of the uncertainties of the exposures due to the variability of the environmental radioactivity. The concerned population was neither warned after the accident nor were there any recommendation for protective measures.

A second goal was to investigate whether the alternative approach to retrospective dosimetry of ^{131}I exposure would work, namely to use ^{137}Cs in soils as a basis to estimate the doses.

Thirdly, we intended to perform a critical comparison of the accuracy of retrospective dosimetry using ^{129}I (or ^{137}Cs). This was possible by comparing our results with thyroid dose estimates based on settlement-averaged results of direct measurements of ^{131}I in human thyroids performed in 1986 (Likhtarov et al., 1994) and evaluated later by Jacob et al. (2006).

Direct measurements of thyroid activities after the Chernobyl accident revealed a high variability of individual exposures compared to the results of deterministic model calculations of the mean thyroid exposures; e.g. Likhtarov et al. (1994). This variability depends – among many behavioural factors – on the small-scale variability of the environmental radioactivity. Since the latter is quantified in this work this will be a first step to assess the uncertainties of thyroid exposure estimates.

The paper is structured in the following way. After describing