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Environmental health risk assessment due to radionuclides and metal(loid)s for Igdir province in Anatolia, near the Metsamor nuclear power plant

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► ***Original article***

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ABSTRACT

Background: There have been growing concerns about environmental pollution and public health issues associated with metal(loid)s and radionuclides. As a baseline for monitorisation, radioactivity and specific metal(loid) levels have been measured to investigate the estimated environmental health risks in Igdir province at the Turkish side of the border, in the vicinity of the Metsamor Nuclear Power Plant, an aging nuclear station without primary containment structures. ***Materials and Methods:*** Concentrations of radioactivity and accumulation of metal(loid)s in the region were measured and hazard quotient, hazard index, and the excess lifetime cancer risk vaues were calculated according to international methods and standards. ***Results:*** Hazard quotient and index values were determined to be lower than the permissible maximum levels. The average estimated excess cancer risk values for terrestrial and cosmic exposures due to radionuclides were calculated as 1.8±0.9E-04 and 2.4±0.04E-04, respectively. The average estimated excess cancer risk value for the radioactivity in water was determined as 15.2±13.6E-05. The mean excess lifetime cancer risk values due to metal(loid) exposure were as Cr<Ni<As in soil and as Cr<Cd<Pb in water. ***Conclusion:*** The mean concentrations of selected metal(loid)s and radionuclide activities were below the maximum permissible limits. Relatively low hazard indices suggest that the population is currently not at any imminent health risk.

Keywords: *Environmental health, cancer, metal(loid), Metsamor, radionuclide, pollution.*

INTRODUCTION

Environmental pollution is a major global health threat. Modern technological developments have brought new challenges concerning environmental safety because unlimited industrialization without proper emission controls and pollution abatement pose a risk to human life [1]. There are increasing ecological concerns about pollution and global public health issues associated with metal(loid)s and radionuclides, which may have harmful effects on human wellbeing through different retention pathways [2,3]. They may cause eco-pollution and health risks, and therefore, exposure to radionuclide materials and certain metal(loid)s are considered objectionable [4,5].

Radioactive substances are defined as materials that contain unstable atoms which produce ionizing radiation through nuclear rearrangement. Radionuclides are radioactive isotopes, and they are widely distributed on earth [6]. Some studies in the relevant literature revealed the health impacts of radiation and showed that radiation exposure might increase cancer risk, and it differs with the dose, age,

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gender, and tissues [7, 8]. Radiation is also accepted as a risk factor for non-cancerous health problems [7]. Radioactivity may cause kidney disorders, cardiovascular diseases, and thus induce hypertension; the mortality risk of cardiac problems increases with dose [9-11]. Risk of stroke increases to some degree, especially at high doses [12].

A metal is a substance with high electrical conductivity, luster, and malleability, which forms cations, and a metalloid is a chemical element that exhibits semimetallic or semiconductor properties [13]. Although it has been diversely defined, the term "heavy metal" usually refers to metal(loid)s having an elemental density >5 gcm^{-3} and an atomic number >20 [14]. The diversity of definitions has led to confusion, and some experts in the field suggested even abandoning the use of the term [14]. Metal(loid)s may accumulate and disturb function in vital organs by binding to cellular components and disrupt function in vital organs [15], or they can remove the essential nutritional minerals from their location and inhibit their bio-action [16]. Metal-toxicity occurs mainly because of their reaction with sulfhydryl enzyme systems [17]. They may cause the production of reactive oxygen species (ROS), and expanded generation of ROS can interfere with fundamental antioxidant defenses of cells and cause oxidative stress [18].

Radioactive contamination and metal pollution in soil and drinking waters can threaten human health in time [19]. Metal(loid)s are likely to attach organic substances to produce organometallic compounds in sediment [20]. Metal(loid)s in the soil can be taken into the body via respiration, ingestion, or skin contact absorption and harm the health [21].

The region potentially carries a critical risk being at the border with Armenia, only ~20 km away from the Metsamor Nuclear Power Plant, an aging Soviet-era nuclear station without primary containment structures (figure 1). Furthermore, the station is built upon an area prone to powerful earthquakes over magnitude 7.0 on the Richter scale, although the plant was not constructed to resist tremors at that level [22,23]. Main pressure parts are worn-out and outdated, and the waste disposal situation is unclear. There is no waste agreement in place, and the emergency response system and evacuation plans are not available [24]. Therefore, the neighboring countries and the European Union are justifiably concerned with environmental health risks [25].

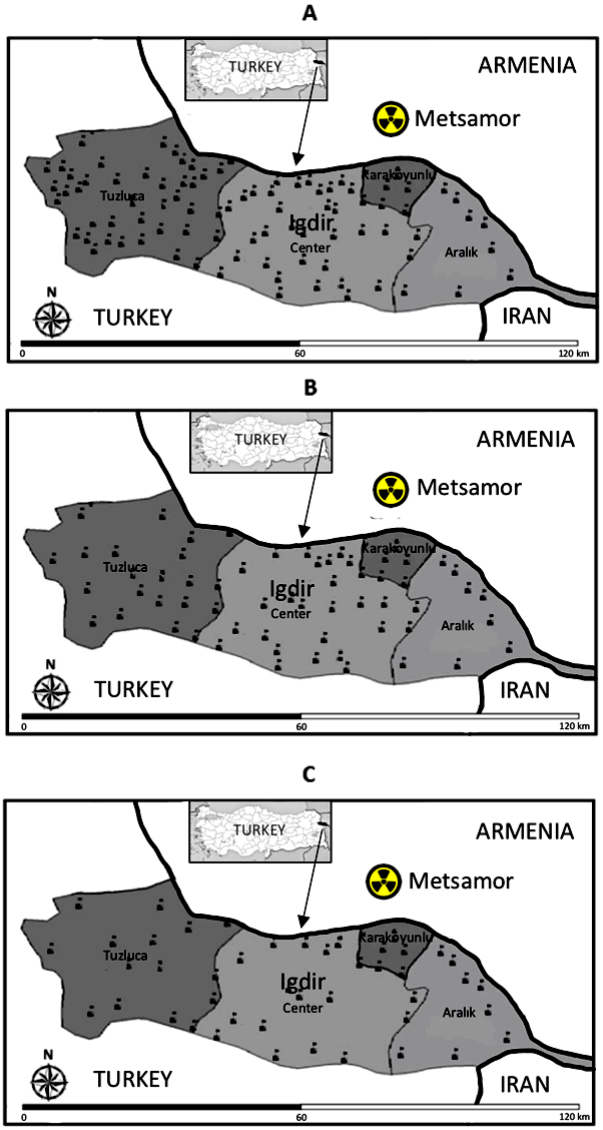


Figure 1. Location of Igdir Province with sampling sites. The region potentially carries a critical risk being at the border with Armenia, only ~20 km away from the Metsamor Nuclear Power Plant, an aging Soviet-era nuclear station without primary containment structures. **A.** Outdoor Absorbed Gamma Dose Rate Measurement Sites **B.** Soil Sampling Sites **C.** Water Sampling Sites.

The present study aims to investigate the risk assessment of health risks due to background

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accumulation of radioactivity and metal(loid)s in soil and water in the natural environment of Igdir province in the vicinity of the Metsamor Nuclear Power Plant. The findings obtained in this study are noteworthy because it can serve as a baseline for future efforts to monitor and evaluate the impacts of the Metsamor Plant, considered to pose a high level of risk.

MATERIALS AND METHODS

For studying the soil radioactivity and terrestrial gamma dose rate, radioactivity in drinking water, outdoor gamma dose rates, and accumulation of metal(loid)s in soil and water, samples were collected from selected sites of Igdir Province. Sampling, sample preparation, and measurements were performed according to international methods and standards [8,26]. Igdir Province is in eastern Turkey, located along the borders with Armenia, Azerbaijan, and Iran (figure 1). Igdir is located in the northeastern part of the country, has an area of approximately 9.587 km2, with a population of 301.766. The province has four districts. The center of the province is situated at an altitude of about 2000 meters over the sea level, and the altitude all over the province changes between 300 to 3300 m. As geographically, plateaus constitute 51% of the city. In some plateaus, which are very wide and undulating, small plains and sediment lakes can be seen. There are many threats to drinking water, which may pose a health risk. Water may come from three primary sources: rainwater, surface water, and groundwater. Water samples in this research were collected as drinking water.

A proportional counter analyzed water samples with the gas flow. The geographic coordinates of the sites were determined by Global Positioning System (GPS). ArcMap module of ArcGIS (10.2 version) mapping software was used for plotting spatial distribution maps for the region. Trace element analysis was determined using x-ray fluorescence (XRF) technique. The powdered samples were passed through a sieve and mixed with eluate agate. The samples were left under a 25-ton hydraulic press for three minutes to pelletize. Then, amounts of elements and compounds were measured in ppm using Wavelength Dispersive X-ray Fluorescence spectrometry. The activity concentration of the radionuclides was determined using a gamma spectrometer. The gamma in-situ measurement system calculated the outdoor gamma dose rates.

Soil radioactivity and terrestrial gamma dose rates, radioactivity in drinking water, outdoor gamma dose rates, and accumulation of metal (loid)s in soil and water were determined. Hazard Quotient (HQ) and Hazard Index (HI) of non-carcinogenic hazards, and the Excess Lifetime Cancer Risk (ELCR) due to metal(loid)s both in soil and drinking water were calculated by equations used as described by the United States Environmental Protection Agency (USEPA) and United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR) [8,26].

Determination of the soil radioactivity and terrestrial gamma dose rate

Soil samples were obtained from the uncultivated locations close to settlements. Open, flat, and undisturbed geographical locations with good water permeability were selected as the sampling points. The foreign bodies were removed, and the remaining soil was placed in clean, sealed, and labeled bags. The samples were dried at 105°C for 24 h, grained, passed through 2 mm sieves, and placed in Marinelli type beakers. The samples were kept one month before the analysis at the airtight condition to allow secular equilibrium between thorium and radium and their decay products. Each sample was counted for 50000 s using a gamma spectroscopy device connected to a coaxial germanium detector, the CANBERRA XtRa in the accredited laboratories of Radioactivity Analysis and Measurement Department in Cekmece Nuclear Research and Training Center (CNRTC). The detector was shielded to reduce the background due to the cosmic rays and the radiation near the system. Full energy peak efficiency was determined using Standard Reference Material prepared by

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the International Atomic Energy Agency [22]. The Minimum Detectable Activity (MDA) value for ^{137}Cs was obtained as 0.5 Bq/kg.

Based on the radioactivity levels of ^{226}Ra, ^{232}Th, ^{40}K, and ^{137}Cs the gamma Absorbed Dose Rate in the Air (ADRA) in nGy h^{-1} at 1 m above the ground level was calculated using the following Equation [8, 26], Equation 1:

$$ADCR = aC_{Ra} \times bC_{Th} \times cC_K \times dC_{Cs} \quad (1)$$

C_{Ra}, C_{Th}, C_K, and $C_{C.s}$ are the Activity Dose Rate Concentrations (Bq kg^{-1}) of ^{226}Ra, ^{232}Th, ^{40}K, and ^{137}Cs, respectively, in the soil sample. The values of a, b, c, and d are coefficients of 0.461, 0.623, 0.0417 and 0.1243 nGy h^{-1} (Bq $kg^{-1})^{-1}$, respectively [8, 26].

Determination of radioactivity in drinking water

The water samples at predetermined sites were transported to the laboratory in 500 cm^3 plastic bottles. Aroutine procedure of sample preparation was used for the radionuclide analyses [27]. After being filtered through a paper, each sample was transferred to a beaker where a smallamount of nitric acid was added to avoid any precipitation on the container walls. After slow evaporation, the sample was moved to a stainless steel counting planchette to be evaporated at a low temperature. Following cooling and weighing for the dry residue, each sample was counted for gross-alpha and gross-beta radioactivities in a low-background proportional counter with the gas flow (Berthold, LB770-PC 10-Channel Low-Level Planchet Counter). The system was commonly used for measuring samples with low natural background radiation. The results were obtained in units of Bq dm^{-3}. The calibration of the low-level counting system used in the measurements was carried out with standard solutions that contained known activities of ^{241}Am for alphas and ^{90}Sr for betas. The following Equation was used to calculate the effective dose (DR_W) due to drinking water radioactivity, Equation 2:

$$DR_W = A_W \times IR_W \times ID_F \times 2 \text{ (for both } \alpha \text{ and } \beta) \quad (2)$$

DR_w is the dose equivalent effective (Sv/year), A_W is Activity (Bq/L), IR_w is the Intake of Water for one person in one year (730 L), and ID_F is the ingestion effective dose equivalent factor for 3.58×10^{-7}Sv/Bq for alpha [28]. ELCR value for 70 years of average life duration was calculated using the following Equation [8, 26], Equation 3:

$$ELCR = DR_W \times DL \times RF \quad (3)$$

DR_w is the annual effective dose equivalent (Sv/year), DL is the Duration of Life (70 years), and RF is a Risk Factor (Sv^{-1}). For risk assessment, the nominal probability coefficient of $7.3\times10^{-2}Sv^{-1}$ recommended by International Commission on Radiological Protection (ICRP) was adopted [29].

Determination of the outdoor gamma dose rates

The outdoor gamma dose rates were determined in the summer. Readings were recorded 1 meter away from the ground at the same sites that soil samples were collected in the four different districts of the region (figure 1). The gamma dose rates were measured by a portable device (Thermo sci.) connected with high sensitivity NaI scintillation detector (NBR model of Thermo sci.) calibrated at the beginning of the study by the accredited Secondary Standard Dosimetry Laboratory (SSDL) of Cekmece Nuclear Research and Training Center (CNRTC). The measurements were performed in the air for two minutes, and the gamma dose rate units were recorded as μR h^{-1} and then converted to nGy h^{-1} using the conversion factor of 8.7 nGy μR^{-1}. The Annual Effective Dose Equivalent (AEDE) was calculated using the following Equation [8, 26], Equation 4:

$$AEDE = ADRA \times DCF \times OF \times T \quad (4)$$

ADRA is Absorbed Dose Rate in the Air (nGy h^{-1}), DCF is the dose conversion factor of 0.7, OF is Occupancy Factor of 0.2, and T is the time (8760 h y^{-1}). ELCR for 70 years of average life duration was calculated using the following Equation [8, 26], Equation 5:

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$$ELCR = AEDE \times DL \times RF \quad (5)$$

DL is the Duration of Life (70 y), and RF is the Risk Factor (Sv^{-1}), fatal cancer risk per sievert. For stochastic effects, RF of ICRP-103 for the public as 0.055 was used [29].

Determination of accumulation of metal (loid)s in soil and water

To determine the number of trace elements in soil, the powdery soil samples were first weighed 12 g, taking care that they were milled below 200 mesh and 3 g of cellulose were added and mixed for five minutes in an agate vat. The samples were pressed for three minutes using a 25-ton hydraulic press with 40 mm diameter steel pellet cups to make pellets. Then, the powder samples prepared were analyzed by Wavelength Dispersive X-Ray Fluorescence spectrometry, and quantities of elements were determined in ppm (µg/g). To determine the number of trace elements in drinking water, calibration standard, and water sample solutions to be analyzed were prepared using 2% HNO3. The solutions were analyzed by Inductively Coupled Plasma-Optical Emission Spectrometry (ICP-OES) (Perkin Elmer Optima 7000 DV) method with autosampler by plotting calibration curves. Additionally, Mercury (Hg) analyzes were performed by ICP-OES/CFHG (Inductively Coupled Plasma - Optical Emission Spectrometry/Continuous Flow Hydride Generation) method. It was provided that the correlation coefficient of the calibration curves was at least r^2 = 0,999. The accuracy of the analysis results was tested with the certified standard reference material named "NIST SRM 1640a - Trace Elements in Natural Water" and quantities of elements were determined in ppm (µg/L). The potential exposure pathways for metal(loid)s in soil, and drinking water are calculated by following the Equations bellow [8, 26], Equation 6:

$$ADI_{S\,ing} = \frac{C \times IR \times EF \times ED \times CF}{BW \times AT} \quad (6)$$

$ADI_{S\,ing}$ is the Average Daily Intake of metal (loid)s ingested from soil (mg/kg-day), C is the concentration of heavy metal (mg/kg) for soil, IR is the Ingestion Rate (100 mg/day) [26], EF is the exposure frequency (350 days/year) [30], ED is the exposure duration (24 years), BW is the bodyweight of the exposed individual (70 kg), AT is the time over which the dose is averaged (365×70=25550 days for carcinogens, 365×ED days for non-carcinogens) [26], and CF is the conversion factor (10^{-6} kg/g), Equation 7:

$$ADI_{S\,inh} = \frac{C \times IR_{air} \times EF \times ED}{BW \times AT \times PEF} \quad (7)$$

$ADI_{S\,inh}$ is the average daily intake of metal (loid)s inhaled from soil (mg/kg-day), C is the concentration of heavy metal in soil (mg/kg), IR_{air} is the Inhalation Rate, 20 m^3/day, PEF is the particulate emission factor, 1.36x109 m^3/kg [26]. EF, ED, BW, and AT are as defined earlier above, Equation 8:

$$ADI_{S\,dems} = \frac{C \times SA \times EF \times ED \times CF}{BW \times AT} \quad (8)$$

$ADI_{S\,dems}$ is the exposure dose from the soil via dermal contact (mg/kg-day), C is the concentration of heavy metal in soil (mg/kg), SA is exposed to the skin area, 3950 cm^2, AF is the soil Adherence Factor in, 0.07 mg/cm^2, ABS is the fraction of the applied dose absorbed across the skin 0.001 [8, 26]. EF, ED, BW, CF, and AT are as defined earlier above, Equation 9:

$$ADI_{W\,ing} = \frac{C \times IR \times EF \times ED}{BW \times AT} \quad (10)$$

$ADI_{W\,ing}$ is the Average Daily Intake of metal (loid)s ingested from water (mg/kg-day); C is the heavy metal concentration in water (µg/L), IRis the daily intake of water, 2.2 L day^{-1} [8, 26], ED is the exposure duration, 70 years [31], EF is the exposure frequency, 365 days/year [32], AT is the time over which the dose is averaged, 365×70 = 25550 days for both carcinogens and non-carcinogens [31], BW is the bodyweight of the exposed individual (70 kg). Non-carcinogenic hazards are characterized by a term called Hazard Quotient (HQ). HQ is a unitless number expressed as the probability of an individual suffering an adverse effect. It is defined as the quotient of ADI or dose divided by the toxicity threshold value, referred to as the chronic

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reference dose (R_{fD}) in mg/kg-day of a specific heavy metal [8, 26], Equation 10:

$$HQ = \frac{ADI}{R_{fD}} \quad (10)$$

For n number of metal(loid)s, the non-carcinogenic effect to the population is a result of the summation of all the HQs due to individual metal(loid)s. Specifically, the total chronic hazard attributable to exposure to all contaminant of potential concerns through a single exposure pathway is known as a Hazard Index (HI)[26]. The Equation below shows the mathematical representation of this parameter for metals(loids) in soil and drinking water [8, 26], Equation 11:

$$HI = \sum_{k=1}^{n} HQ_k = \sum_{k=1}^{n} \frac{ADI_k}{R_{fDk}} \quad (11)$$

HQ_k, ADI_k, and R_{fDk} are values of heavy metal k. For carcinogens, the risks are estimated as the incremental probability of an individual developing cancer over a lifetime as a result of exposure to the potential carcinogen. The Equation for calculating the Excess Lifetime Cancer Risk (ELCR) due to metal(loid)s both in soil and drinking water is [8, 26], Equation 12:

$$ELCR = \sum_{k=1}^{n} ADI_k \times SF_k \quad (12)$$

Excess Lifetime Cancer Risk values is a unitless probability of an individual developing cancer over a lifetime. ADI_k(mg/kg/day) and SF_k $(mg/kg/day)^{-1}$ are the average daily intake and the cancer slope factor, respectively, for the kth heavy metal, for n number of metal(loid)s. The slope factor converts the estimated daily intake of the heavy metal averaged over a lifetime of exposure directly to the incremental risk of an individual developing cancer [8, 26].

In summary, descriptive statistics are given in tables as mean ± standard deviation (SD). Categorical variables are summarized as numbers. Statistical analyses were evaluated using the IBM SPSS Statistics software v20.0 (IBM Corp., Armonk, NY, USA).

RESULTS

A total of 113 soil samples was collected from predetermined sites and then analyzed. Radionuclide concentrations (Bq/kg) for ^{40}K, ^{226}Ra, ^{232}Th and ^{137}Cs in soil samples were 441±182, 13±12.6, 14.1±5.2, and 16.0±6.0, respectively. Mean Absorbed Dose Rate values in Air (nGy/h) was meusured for outdoor as 82.4±18.1, and calculated for outdoor and cosmic as 35.2±12.7 and 47.3±16.3, respectively. Radioactivity as gross α activity and gross β activity in water were 0.053±0.04 and 0.19±0.13, respectively (table 1).

The average annual effective dose values (mSv) resulting from terrestrial and cosmic gamma radiations were calculated as 0.04±0.02 and 0.06±0.02, respectively. Cosmic radiation exposure dose levels primarily depend on the altitude of the area. Accordingly, the average estimated excess cancer risk values were calculated as 1.78±0.88E-04 and 2.39±0.04E-04 for terrestrial and cosmic exposures, respectively. Annual cumulative biologic effective dose due to radiologic exposure that arises from radioactivity in drinking water in the region was determined as 27.83±24.81μSv. Accordingly, the average estimated excess cancer risk value was determined as 15.24±13.6E-05 (table 2).

The collected 113 soil and 43 water samples were also analyzed to find out health risks depending on metal(loid) accumulation in the research region. Mean metal(loid) concentration of Cr, Mn, Ni, Cu, Zn, As, and Pb in soil were determined as 222 (27-1235), 979 (550-2012), 128 (21-592), 52 (30-96), 112 (59-422), 11 (4-25), and 12 (7-34) μg/g, respectively and mean metal(loid) concentration of Cd, Cr, Cu, Ni, Pb, Zn, and Ba for Cd, Cr, Cu, Ni, Pb, Zn, and Ba in drinking water were determined as 0.3 (LDL-1.1), 2.0 (LDL-9.1), 2.1 (LDL-25.8), 1.7 (LDL-3.8), 1.2 (LDL-4.2), 67.1 (LDL-2020), and 35.3 (1.5-135.7) μg/L, respectively. The amount of metal (loid) concentration in water is related to interactions of water sources and their

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pathways. Therefore, distribution of metal (loid) s in water can be different from metals in soil (table 3).

Reference dose (Rfd) values and cancer slope factors (SF) for each metal(loid) were given in table 4. The non-carcinogenic hazard quotient (HQ) values were determined for Cr, Ni, As, Pb, Zn, Mn, and Cu in soil as 1.36E+00, 1.69E-02, 8.06E-02, 1.25E-02, 6.22E-04, 3.75E-02, and 3.25E-03, respectively and for Cd, Cr, Cu, Ni, Pb, Zn, and Ba in water as 4.79E-01, 2.38E-01, 2.39E -02, 2.74E-02, 1.52E-01, 4.20E-02, and 5.56E-02, respectively. The mean excess lifetime cancer risk values due to metal(loid) exposure were as Cr<Ni<As in soil and as Cr<Cd<Pb in water (table 5).

Table 1. Radionuclide concentrations in soil, absorbed gamma dose rates and radioactivity in water.

District	Radionuclide Concentration (Bq/kg)				Absorbed Dose Rate in Air (nGy/h)			Radioactivity in Water (Bq/L)	
	^{226}Ra	^{232}Th	^{40}K	^{137}Cs	Outdoor (measured)	Outdoor (calculated)	Cosmic (calculated)	Gross α Activity	Gross β activity
Aralık	426±133	6.3±3.7	14.2±4.4	13.9±3.5	85.4±17.5	**31.2±6.2**	**54.2±18.0**	0.069±0.045	0.23±0.10
Center	436±148	11.5±7.5	15±4.8	18.4±5.0	87.1±16.8	**35.2±8.9**	**51.9±14.8**	0.055±0.044	0.20±0.13
Karakoyunlu	345±96	8.5±6.5	13.1±4.9	13.0±4.6	73.5±18	28.1±9.4	45.4±10.6	0.089±0.051	0.28±0.12
Tuzluca	486±231	18.7±16.7	13.8±6.1	16.5±7.2	81.1±18.8	39.6±16.3	41.5±16.7	0.025±0.035	0.10±0.07
Igdir	441±182	13±12.6	14.1±5.2	16.0±6.0	82.4±18.1	35.2±12.7	47.3±16.3	0.053±0.047	0.19±0.13

Average Values ± Standart Deviation

Table 2. Health risks (ELCR) due to background radioactivity.

District	Terrestrial		Cosmic		Radioactivity in Water	
	Annual Effective Dose (mSv)	Excess Lifetime Cancer Risk	Annual Effective Dose (mSv)	Excess Lifetime Cancer Risk	Annual Effective Dose (μSv)	Excess Lifetime Cancer Risk
Aralık	0.03±0.01	1.42±0.54E-04	0.06±0.01	2.30±0.37E-04	12.81±18.14	7.01±9.93E-05
Center	0.04±0.01	1.58±0.53E-04	0.07±0.02	2.74±0.36E-04	28.59±23.07	15.65±12.63E-05
Karakoyunlu	0.04±0.01	1.78±0.67E-04	0.06±0.02	2.63±0.18E-04	35.98±23.54	19.7±12.89E-05
Tuzluca	0.05±0.02	2.00±1.11E-04	0.05±0.02	2.1±-0.16E-04	46.57±26.44	25.5±14.48E-05
Igdir	0.04±0.02	1.78±0.88E-04	0.06±0.02	2.39±0.04E-04	27.83±24.81	15.24±13.6E-05

Average Values ± Standart Deviation

Table 3. Average concentration of certain metal(loid)s in soil and drinking water.

Soil							
District	Metal(loid) Concentration (μg/g)						
	Cr	Mn	Ni	Cu	Zn	As	Pb
Aralık	247 (27-373)	863 (550-1070)	136 (21-218)	50 (35-64)	87 (59-193)	15 (10-25)	12 (7-34)
Center	196 (70-327)	894 (616-1181)	127 (45-218)	51 (30-70)	112 (63-322)	12 (6-20)	13 (10-26)
Karakoyunlu	312 (91-464)	960 (639-1156)	167 (36-243)	50 (37-55)	76 (65-92)	13 (12-15)	10 (8-12)
Tuzluca	199 (60-1235)	1101 (642-2012)	112 (39-592)	55 (35-96)	137 (70-422)	8 (4-13)	14 (8-22)
Igdir	222 (27-1235)	979 (550-2012)	128 (21-592)	52 (30-96)	112 (59-422)	11 (4-25)	12 (7-34)
Drinking Water							
District	Metal(loid) Concentration (μg/L)						
	Cd	Cr	Cu	Ni	Pb	Zn	Ba
Aralık	0.7 (LDL-1.1)	3.2 (0.3-6)	4.2 (LDL-14.9)	1.9 (1.2-2.7)	1.3 (LDL-1.8)	211.1 (3.5-2020)	42.6 (1.5-60.2)
Center	0.1 (LDL-0.7)	2 (LDL-8.9)	1.4 (LDL-10.2)	1.8 (0.5-2.4)	1.2 (LDL-2.7)	16.6 (LDL-177.4)	40.6 (2.2-135.7)
Karakoyunlu	0.7 (LDL-1.1)	3.6 (0.2-9.1)	3.4 (LDL-25.8)	2.2 (1.3-2.8)	0.2 (LDL-1)	72.0 (0.4-354.2)	67 (39.1-97.8)
Tuzluca	0.1 (LDL-0.8)	0.5 (LDL-1.1)	0.9 (LDL-5.2)	1.2 (LDL-3.8)	1.8 (LDL-4.2)	28.8 (LDL-411.8)	9.8 (1.9-24.9)
Igdir	0.3 (LDL-1.1)	2.0 (LDL-9.1)	2.1 (LDL-25.8)	1.7 (LDL-3.8)	1.2 (LDL-4.2)	67.1 (LDL-2020)	35.3 (1.5-135.7)

LDL is for lower than detection limits.

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Table 4. Reference dose values (Rfd) and cancer slope factors (SF). [26, 31, 59]

Metal(loid)	R_{fD} (mg/kg-day)			SF (mg/kg/day)$^{-1}$		
	Oral	Inhalation	Dermal	Oral	Inhalation	Dermal
Cr	3.00E-03	2.86E-05	6.00E-05	5.01E-01	4.20E+01	2.00E+01
Ni	2.00E-02	2.06E-02	5.40E-03	1.70E+00	8.40E-01	4.25E+01
As	3.00E-04	3.00E-04	1.23E-04	1.50E+00	1.51E+01	1.50E+00
Pb	3.50E-03	3.52E-03	5.25E-04	8.50E-01	NA	NA
Cd (W)	5.00E-04	1.00E-03	1.00E-05	1.50E+01	NA	NA
Cd (S)	1.00E-03	1.00E-03	1.00E-05	3.80E-01	6.30E+00	3.80E-01
Ba	2.00E-01	NA	NA	NA	NA	NA
Zn	3.00E-01	3.00E-01	6.00E-02	NA	NA	NA
Mn	1.40E-01	1.43E-05	2.33E-02	NA	NA	NA
Hg	1.00E-04	8.57E-05	2.10E-02	NA	NA	NA
Cu	4.00E-02	4.20E-02	1.20E-02	NA	NA	NA

^{W} is for metal(loid) in Water, ^{S} is for metal(loid) in Soil, NA: Not Available.

Table 5. Health risk due to metal(loid) concentration in soil and drinking water.

Soil										
District	Hazard Quotient (HQ)							Excess Lifetime Cancer Risk		
	Cr	Ni	As	Pb	Zn	Mn	Cu	Cr	Ni	As
Aralık	1.51E+00	1.79E-02	1.10E-01	1.25E-02	4.92E-04	3.32E-02	3.13E-03	1.85E-03	2.28E-03	3.85E-05
Center	1.91E+00	2.22E-02	9.52E-02	1.04E-02	4.30E-04	3.72E-02	3.19E-03	8.58E-04	2.83E-03	3.34E-05
Karakoyunlu	1.20E+00	1.67E-02	8.79E-02	1.35E-02	6.33E-04	3.44E-02	3.19E-03	1.47E-03	2.13E-03	3.08E-05
Tuzluca	1.20E+00	1.46E-02	5.86E-02	1.35E-02	7.52E-04	4.22E-02	3.44E-03	5.39E-04	1.86E-03	2.05E-05
Igdir	1.36E+00	1.69E-02	8.06E-02	1.25E-02	6.22E-04	3.75E-02	3.25E-03	1.67E-03	2.16E-03	2.82E-05
Drinking Water										
District	Hazard Quotient (HQ)							Excess Lifetime Cancer Risk		
	Cd	Cr	Cu	Ni	Pb	Zn	Ba	Cd	Cr	Pb
Aralık	5.16E-01	3.38E-01	3.62E-02	2.94E-02	1.25E-01	2.22E-01	6.71E-02	3.87E-03	5.07E-04	3.72E-06
Center	-	2.67E-01	1.39E-02	2.83E-02	1.38E-01	2.32E-02	6.39E-02	-	4.00E-04	4.10E-06
Karakoyunlu	4.79E-01	3.75E-01	4.46E-02	3.42E-02	-	7.55E-02	1.05E-01	3.59E-03	5.62E-04	-
Tuzluca	-	5.88E-02	1.36E-02	2.14E-02	1.90E-01	3.55E-02	1.54E-02	-	8.82E-05	5.65E-06
Igdir	4.79E-01	2.38E-01	2.39E-02	2.74E-02	1.52E-01	4.20E-02	5.56E-02	3.59E-03	3.57E-04	4.52E-06

DISCUSSION

In this study, the integral health risks due to two crucial environmental pollutants, radionuclides, and metal(loid)s were investigated for Igdir province. Radiation exposure is accepted as a risk factor for cancerous and non-cancerous health problems [7-12]. EPA has classified As and Hexavalent Cr as Group A-human carcinogen, Cd, and Pb as a Group B -probable human carcinogen, while Cu is accepted as Group D - not classifiable as to carcinogenicity in humans [26,33]. Concerning carcinogenic health risk, excess lifetime cancer risk values were determined using equation 12. It was determined that the estimated mean excess lifetime cancer risk values due to metal (loid) exposure were as Cr<Ni<As in soil and as Cr<Cd<Pb in water.

HQ and HI values due to metal(loid) exposure were determined to be lower than the permissible maximum values for both soil and drinking water. Thus, the soil and the water of the region are unpolluted with metal(loid)s. Relatively low hazard indices suggest that the population is currently not at any imminent health risk because of metal(loid)s. Table 6 presents background radiation studies conducted in various other regions, and table 7 presents environmental metal(loid) studies

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conducted in different cities around the world.

The determined average values of radionuclide and gross alpha and beta mean activities for the region were in the ranges of the values found in other regions of Turkey [28,34-36, 42,43]. However, the average value of ADRA for

Table 6. Background radioactivity studies carried out in different regions. [27, 28, 35, 36, 38-44]

Referance	Location	ADRA (nGy/h^{-1})	Activity in Soil (Bq/kg)				Activity in Water (mBq/L)	
			^{40}K	^{226}Ra	^{232}Th	^{137}Cs	Gross-α	Gross-β
Taşkın *et al.* [38]	Kırklareli, Turkey	118	667	37	40	8	-	-
Karahan *et al.* [27]	Istanbul, Turkey	65	342	21	37	-	23	70
Karahan *et al.* [35] **Taşkın *et al.*** [36]	Bursa, Turkey	90	430	60	-	5	69	67
Kapdan *et al.* [40]	Balikesir, Turkey	127	675	-	55	3	-	-
Kapdan *et al.* [39]	Çankırı, Turkey	70	357	-	22	4	250	260
Değerlier *et al.* [41] **Değerlier *et al.*** [42]	Adana, Turkey	67	298		21	7	10	86
Kobya *et al.* [28] **Kucukomeroglu *et al.*** [43]	Artvin, Turkey	-	358	-	19	54	46	91
Zhang 2017 [57]	Xitulvye, China	78	396	49	63	-	-	-
Khan *et al.* [44]	North Waziristan, Pakistan	-	453	69	123	-	-	-
WHO, [37]		-	-	-	-	-	500	1000
Worldwide Average (UNSCEAR 2018) [8]		54	400	35	30	-	-	-
Present Study	Igdir, Turkey	82	441	13	14	16	53	190

Table 7. Metal(loid) studies carried out in different cities around the world [19, 21, 45-56].

Reference	Location	Soil (µg/g)						
		Cr	Mn	Ni	Cu	Zn	Pb	As
De Miguel *et al.* [45]	Madrid, Spain	75	437	14	72	210	161	-
Lux *et al.* [46]	Hamburg, Germany	95	750	63		516	218	-
Hu *et al.* [47]	Nanjing, China	12	32	16	29	61	60	48
Chen *et al.* [19]	Hong Kong	-	-	-	16	59	90	17
Navas *et al.* [48]	Aragon, Spain	21	320	19	10	56	8	12
Wei *et al.* [21]	Zhejiang, China	48	-	-	-	-	69	16
Ruiz-Cortes *et al.* [49]	Sevilla, Spain	41	368	22	41	121	146	-
Present Study	Igdir, Turkey	222	979	128	52	112	12	11
Reference	**Location**	**Drinking-Water (µg/L)**						
		Cr	**Ba**	**Ni**	**Cu**	**Zn**	**Pb**	**Cd**
Waseem *et al.* [50]	Sialkot, Pakistan	0,030	-	0,10	0,06	0,2	0,49	-
Waseem *et al.* [50]	Karachi, Pakistan	0,012	-	0,04	0,12	-	0,01	-
Turdi *et al.* [51]	Dawanqi, China	3,790	-	0,86	-	4,5	0,04	0,008
Turdi *et al.* [51]	Keyiri, China	2,520	-	1,22	-	1,8	0,05	0,016
Huseyinli *et al.* [52]	Oguz–Gabala PL, Azerbaijan	0,530	110	1,89	3,89	14,2	1,63	0,570
Ahmad *et al.* [53]	Sungai Petani, Malaysia	2,190	-	5,63	-	-	5,18	2,810
Bajwa *et al.* [54]	SW-Punjab, India	28,300	-	35,00	145	833	46,0	-
Arain *et al.* [55]	Bannu, Pakistan	-	10046	1,73	10	235	-	-
Kruawal *et al.* [56]	Bangkok, Thailand	-	43	0,3	-	250	-	-
Ahmad *et al.* [58]	Kulim, Malaysia	0,1	-	0,59	-	-	0,56	0,31
USEPA [26]		100	-	-	-	-	15	5
WHO, 2011 [37]		50		70			10	3
Present Study	Igdir, Turkey	2,270	35	1,74	2,10	67	1,20	0,300

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the research region is quite high [8] and that could be explained by the effects of cosmic radiation due to the higher altitude of the region compared to other regions in the country. Moreover, it was seen that beta activity was higher than the values in other cities; on the contrary, the alpha activity in the region was respectively has lower values. This should be the result of the positive relation between alpha activity with the radium and thorium activities (e.g., Adana versus Igdir).

When the metal(loid) concentrations determined for soil were compared with the values of the other cities, it is seen that the concentrations of non-carcinogenic metals in the region are higher than those of the investigated cities. However, values for carcinogen heavy metals such as As and Pb are lower. This might be related to that a large part of Igdir consists of rural areas, and that the soil structure of rural areas has rich geochemical characteristics. However, this situation is entirely different for the concentration of the metal(loid)s in the water. The determined radionuclide concentrations in soil and alpha-beta activity in water were in the range of studies carried out in other regions in Turkey and close to worldwide averages. However, as expected, the average outdoor GDR determined for the region has the highest value determined in Turkey, which arises from the cosmic radiation effect due to the very high altitude of the region. Besides, alpha-beta activity and concentration of all metal(loid)s in water were found lower than the permissible maximum limits specified by WHO [37].

In the present investigation, it was observed that all parameters for radiation hazards were lower than the permissible maximum values. Thus, the soil of the studied area can be used by the residents without any serious radiological hazards. Radiation exposure may lead to an increased incidence of cancer. Exposed individuals should be studied for an extended period. Cancer risk increases due to exposure to moderate and high doses of radiation. However, our knowledge of the risk by acute low doses or low dose rates is limited. Therefore, the dose-response relation needs to be investigated by further molecular epidemiological studies in the future.

This is the first report on the environmental health risk assessment due to pollutants of radionuclides and metal(loid)s for the Turkish side of the border, in the vicinity of Metsamor Nuclear Power Plant, which has a risky condition. The emergency response system and evacuation plans are needed urgently. Based on the measurements of this study, *currently*, there are no health risks to the public for using the water and the soil of the region. Further studies should address the other kinds of pollutants as well for preventing or minimizing the harm from environmental pollution. More measures should be taken to control pollution and protect public health. The authors hope the findings obtained in this research will be useful for further studies to assess the doses of radioactivity and metal (loid) concentrations, which would help to formulate regional regulations for permissible levels.

CONCLUSION

Igdir Province of Turkey is only ~20km away from the Metsamor Nuclear Power Plant in Armenia. Measured levels of background radioactivity, as well as the concentration of radionuclide and metal(loids) in the soil and drinking water samples, indicate that currently, there is no health risk in the region. This study may become a valuable baseline for future research that aims to monitor the impacts of this aging plant built with Soviet-era technology. Attention should also be paid to radionuclide and metal(loid)s pollution in the water and soils for the safety and health of the local population. Although metal(loid)s in the soil samples did not seem harmful to human beings, monitorisation and protective measures must be taken to protect the environment. Primary healthcare professionals are uniquely suited to take a leadership role in the education of the public on the issues of environmental pollution and protection from radiation and metal(loid) exposure.

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