## Regular article

# Regional characteristics of 40 elements in water around Chornobyl nuclear power plant after some decades since the accident

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## **Abstract**

Concentrations of 40 kinds of elements in water samples collected around Chornobyl nuclear power plant (CNPP) in September 2017 were determined using ICP-MS to investigate possible adverse effects on residents' health. Regional characteristics of elements of Chornobyl and Narodychi district, which suffered high radioactivity due to the accident on 1986, were compared with those of other cities in Ukraine, Belarus, and Japan. The concentrations of toxic elements such as As, Cd, Pb, and Se in Chornobyl and Narodychi were lower than regulation values of WHO guidelines for drinking-water quality, and not particularly higher than those of other cities. Among three well water samples of Chornobyl, one or two contained much higher concentrations of Mn, Zn or La than tap water or hand washing water. Three well water and one tap water samples of Chornobyl were also collected in April 2018, August 2019, and January 2020 in order to know the yearly change of elements. Changes in concentrations of alkali and alkaline earth elements were relatively small, and those of transition elements were large. Pb concentrations of most of samples were lower than 0.3 μg/L, but an extremely high value of 340 μg/L was observed temporarily in one well water sample in 2019, although the source of Pb was not clear. These results did not suggest notable health problems associated with elements in the water of Chornobyl.

**Keywords:** Drinking water quality, Metal elements, Chornobyl, ICP-MS

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## **Introduction**

No one can live without water. Adults need about 2.5 liters of water a day, of which two liters are taken as drinks such as simple water, tea, coffee, juice, and so on. Most people consume simple water, for example, tap water, well water, or bottled water. The developed countries have water supply systems and serve hygienic drinking water to residents. Even in the developed countries, however, municipal water pipes do not reach secluded places in



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the mountain areas. In developing countries, there are many cases in which rural areas do not always have water supply systems. The residents living around these areas use nearby water such as well water, surface (river or pond) water or rain water. These waters are not always tested regarding security for health. Natural water contains various minerals coming from environmental conditions or chemical contaminants caused by agricultural or industrial residues. WHO has established criteria of drinking water in order to prevent negative effects of human health caused by drinking water  $[1]$ . In our research, we have been paying attention to water quality in terms of its elements. We collected water samples and analyzed various elements, then considered their health effects. Previously, we reported the results of water samples collected in the People's Republic of Bangladesh with arsenic contamination from groundwater, and the Republic of Kazakhstan with environmental problems from the Aral Sea [2].

In the present study we paid attention to Ukraine and Belarus, which suffered environmental destruction from radioactive materials generated by explosions of the Chornobyl Nuclear Power Plant (CNPP) about 30 years ago. We visited Ukraine and Belarus in September, 2017. We obtained permission to get into the 30 km area of the CNPP in advance. In Ukraine we collected water samples in the CNPP area and in the Narodychi Raion (district) of Zhytomyr Oblast, located 50 - 70 km south-west from CNPP, known for its high radioactivity due to the accident of 1986, and also in the capital, Kyiv. The southern Belarus area was heavily exposed to radioactivity by the accident of CNPP. We also collected water samples in the capital, Minsk, and Brest, in south-west Belarus. For comparison, tap water and bottled water of Japan were also analyzed. We went to Ukraine again in April, 2018, August, 2019, and January, 2020, and collected well water that was used for daily life by residents within 10 - 30 km of CNPP and tap water of a guest house in Orane village of Chornobyl. We focused on regional characteristics of element concentrations of water around CNPP. Concentrations of 40 kinds of elements in water were determined and we discuss their effects on residents' health,

## **Materials and Methods**

#### **Water samples**

Water samples were collected in polypropylene containers which were pre-cleaned by soaking in 35% nitric acid solution for 2 days and rinsed with ultrapure water (18.2 M $\Omega$ /cm).

The water samples of 2017 (n=37) were divided into 5 groups according to the water type. a) Mineral water; bottled drinking water with trademark of "mineral water". b) Drinking water; water in containers provided for drinking in commercial products. c) Tap water; supplied by the local governments; users use it as drinking water. d) Well water: usual well water without any specialized treatment. e) Hand-washing water; piped water supplied at toilets or wash rooms for hand washing. The numbers and regions of samples are listed in Table 1.

#### **Methods**

Water samples were passed through 0.45 μm filter membranes, and then diluted 5 times with 0.5% nitric acid prepared with 68% HNO3 (ultrapure grade; Tama Chemicals Co., Kawasaki, Japan) and ultrapure water. Twenty μg/L Tb was added as an internal standard. The elemental concentrations in each diluted





solution were determined using an inductively coupled plasma mass spectrometer (ICP-MS; Agilent 8800; Agilent Technologies, Tokyo, Japan). As standard solutions, 0, 0.4, 4.0, and 40.0 μg/L of the multi-element standard solutions XSTC-1 and XSTC-13 (SPEX Industries Inc., NJ, USA) diluted with 0.5% nitric acid were used. The concentration of 40 elements was calculated using standard calibration curves with the signal intensity of Tb as the internal standard. Certified reference materials (JSAC-0301-3b, JSAC-0302-3b, JSAC-0302-3c, Japan Society for Analytical Chemistry) were analyzed for assessing the validity of measurements.

Operation conditions of ICP-MS were as follows. RF power: 1550 W, RF matching 1.50V, sampling position: 10 mm, plasma gas flow: 15 L/min. carrier gas flow: 1.03 L/min. For 2017 samples,  ${\rm ^7Li,}$   ${\rm ^8}$ <sup>135</sup>Ba, <sup>139</sup>La, and <sup>193</sup>Ir were measured by no gas mode, and <sup>23</sup>Na, <sup>52</sup>Cr, <sup>55</sup>Mn, <sup>59</sup>Co, <sup>60</sup>Ni, <sup>63</sup>Cu, <sup>66</sup>Zn, <sup>74</sup>Ge, <sup>75</sup>As, <sup>78</sup>Se, <sup>88</sup>Sr, <sup>98</sup>Mo, <sup>103</sup>Rh, <sup>107</sup>Ag, <sup>118</sup>Sn, <sup>205</sup>Tl, <sup>208</sup>Pb, <sup>209</sup>Bi, <sup>232</sup>Th  $H_2$ , and <sup>40</sup>Mg, <sup>44</sup>Si, <sup>47</sup>P, <sup>32</sup>S, and <sup>60</sup>Ca were measured using  $O_2$  as reaction gas at 7 and 0.3 mL/min, respectively. These samples were also measured by no gas mode, and the results were compared with those obtained by collision or reaction gas mode. The concentrations measured by different mode coincided with each other although the sensitivities to several elements by no gas mode were lower, indicating the differences in analysis mode did not affect the results. Measurement of samples collected in 2018, 2019, and 2020 were performed by no gas mode.

Instrument detection limits were defined as the concentrations 3-times the standard deviations (n=10 times measurements) of signal intensities of 0.5% nitric acid. The water hardness was defined as the amounts of  $CaCO<sub>3</sub>(mg/L)$ , which was calculated from amounts of Ca and Mg obtained by ICP-MS. Molecular weight of CaCO<sub>3</sub> is 100.1, atomic weights of Ca and Mg are 40.1 and 24.3, respectively. The hardness is, therefore, calculated as Ca (mg/L)  $\times$  2.5 + Mg (mg/L)  $\times$  4.1.

#### **Results**

Analytical results of element concentrations of water samples of 2017 are listed by region in Table 2. Results of nine elements and one index of individual samples are indicated in Figures 1A -1J.

#### **1. Group 1 elements (alkali metals)**

Among alkali metals (Li, Na, K, Rb and Cs) in Table 2A, the concentration of Li was 0.06 - 25.13 μg/L, where the average values of Chornobyl and Narodychi district were a little higher than those in other areas. Average concentrations of Na showed similar area dependencies to Li, and individual differences of samples were significant as shown in Figure 1A. The highest concentration of 41 mg/L was found in the mineral water bought from Minsk railroad station, the second highest was drinking water from Chornobyl, and the third highest was the tap water from Narodychi district. Average concentration of K in Chornobyl samples was higher than the other areas. Among Chornobyl samples, two kinds of well water showed especially high levels of K (Figure 1B). Rb concentrations were low; several μg/L or lower. Cs was scarcely detected in all samples.

## **2. Group 2 elements (Alkaline earth metals)**

Of the alkaline earth metals, concentrations of Be were lower than detection limit (0.07 μg/L) except for one well water C of 0.08 μg/L. The highest concentration of Mg was observed in Narodychi district, and average concentration of Minsk was followed by the other four regions. The highest concentration of Ca was also observed in Narodychi district, which was about 2 or 3 times the average concentrations in Chornobyl, Kyiv, Minsk, and Brest, and about 4 times that of Tokyo. The concentrations of Mg and Ca of individual specimens, and hardness calculated as CaCO<sub>3</sub> are shown in Figure 1C, 1D and 1E. Concentrations of Sr in Chornobyl and Narodychi district were a little higher than in Kyiv and Brest, and much higher than in Minsk and Tokyo. Concentrations of Ba were less than 120 μg/L, except for one sample of mineral water from Minsk of 773 μg/L.

## **3. Group 3-12 elements (transition elements)**

The results of transition elements are shown in Table 2B. Zirconium (group 4) concentrations were lower than 0.2 μg/L, except for well water B in Chornobyl (1.70 μg/L). Average concentration of V (group 5) was more than 1.0 μg/L in Chornobyl and Tokyo, but was less than 0.25 μg/L in other regions. Average concentrations of total Cr (group 6) in each area were from 0.08 to 0.37 μg/L, and the maximal value was 1.12μg/L in well water B of Chornobyl, which was much lower than the 50 μg/L of the WHO guideline value [1]. Molybdenum (group 6) concentrations were lower than 1.14 μg/L. Health-based guideline

value of water has not been proposed for Mo.

Average concentration of Mn (group 7) of Chornobyl, 33 μg/L, was higher than that of other regions (Table 2B). As shown in Figure 1F, Mn concentrations of two samples, hand-washing B of Chornobyl and tap water of Narodychi district, were higher than the 80 μg/L of the WHO guideline [1]. Mn concentrations in other samples were lower than 10 μg/L, except for well water A in Chornobyl (44.2 μg/L), hand-washing waters D, L, and R in Kyiv, Minsk, and Brest (21.6, 10.8, and 13.8 μg/L). Iron (group 8) concentrations were lower than 2 μg/L, except for one well water of 53 μg/L which was much lower than that in Japan and that of the USEPA criterion of 300 μg/L [3, 4]. Cobalt (group 9) concentrations were lower than 0.8 μg/L. Rhodium (group 9) was detected in several samples at 2.9-12.8 μg/L. Iridium (group 9) was under detection limit in all samples. Nickel (group 10) concentration was 10.76 μg/L in Narodychi district and average concentration was 2.13 μg/L in Chornobyl. These values were higher than in other regions, but were much lower than the 70  $\mu$ g/L of the WHO guideline [1].

Copper (group 11) was found in almost all samples, and average concentrations of each region were from 3 to 33 μg/L. The highest concentration of 125 μg/L was much lower than the 2000 μg/L of the WHO criterion [1], 1000 μg/L of Japan [3], and 1300 μg/L of USEPA [4]. Silver (group 11) concentrations were lower than 0.120 μg/L, except for the 2.69 μg/L of drinking water in Kyiv. The concentrations of Zn (12 group) of Chornobyl and Narodychi district were higher than those in the other regions. As shown in Figure 1G, the concentrations of Zn in most samples were lower than 400 μg/L, except for the 4830 μg/L of well water A of Chornobyl , 2122 μg/L of hand-washing water L of Minsk, 1382 μg/L of hand-washing S of Brest and 948 μg/L of Narodychi district. Cd (group 12) concentrations were lower than 0.24 μg/L in the tap water of Narodychi district, which was much lower than the WHO criterion of  $3 \mu g/L$  [1].

## **4. Group 13-16 (base metal and nonmetal)**

Analytical results of base metal (Group 13 -15) and nonmetal (Group 14-16) are shown in Table 2C. Aluminum (group 13) concentrations were very low in Narodychi district, Brest and Tokyo, 3 μg/L or lower, and varied from under detection limit to 150 μg/L in Chornobyl, Kyiv and Minsk. Indium (group 13) and Tl (group 13) concentrations were scarcely detected or under detection limit in all samples. Germanium (group 14) was detected in most samples, however, the concentrations were very low, less than 0.2 μg/L. Tin (group 14) concentrations were under detection limit except in 2 samples. Lead (group 14) was detected in many samples but the concentrations were low, as shown in Figure 1H. There were only 2 samples with the concentration more than 1 μg/L, and the highest concentration of 1.47 μg/L in the hand washing water I of Minsk was much smaller than the WHO criteria of 10 μg/L [1]. Antimony (group 15) concentrations were lower than 1.4 μg/L (guideline value is 20 μg/L). Bismuth (Group 15) concentrations of the samples were very low, 0.05 μg/L or less.

Silicon (group 14), one of nonmetal elements existing abundantly in the Earth's crust, was detected in all samples from 108 to 14,863 μg/L. The concentrations of P (group 15) were lower than 100 μg/L, except for 650 μg/L of well water C. Arsenic (group 15) concentrations were from under detection limit (<0.003) to 1.82 μg/L and were much lower than the 10 μg/L of the WHO criterion [1]. Sulfur (group 16) concentrations were lower than 300 μg/L, except for about 1,200 μg/L in tap water of Narodychi district. Selenium (group 16) concentrations of all samples were lower than 0.8 μg/L, much lower than the 40 μg/L of the WHO criterion [1].

## **5. Lanthanoid and actinoid elements**

The results of lanthanoid and actinoid elements are shown in Table 2C. The average concentration of La was high in Chornobyl. As shown in Figure 1I , La was detected in well water B and C at about 18,000 μg/L, and in the well water A at 693 μg/L. La concentrations of all other samples were lower than 130 μg/L. Among actinoid elements, Th was scarcely detected in all samples, however, U was detected, and the concentrations were different depending on the area. As shown in Figure 1J, U concentrations of Chornobyl, Narodychi district, Brest and Tokyo were very low, from under detection limit to 0.12 μg/L, and most samples of Minsk and two samples of Kyiv were from 0.2 to 1.5 μg/L.

#### **6. Yearly changes in elemental concentrations**

We observed yearly changes in element concentrations of three well waters and tap water of Chornobyl from 2017 to 2020. As shown in Figure 2, changes in concentrations of alkali and alkaline earth elements were relatively small, and those of transition

elements were large. Concentrations of Mn in well water A changed from under detection limit to 126 μg/L, and those of Zn from 69 to 3240 μg/L. Manganese in well water B and C also changed from under detection limit to 0.82 μg/L, Zn in well water B and C changed from 15 to 85 μg/L .

Lead concentrations of most samples were lower than 0.3 μg/L, however, an extremely high value of 340 μg/L was observed in well water A of 2019. Lead concentration of the next year, 2020, was reduced to 0.3 μg/L again.

#### **Discussion**

The results of determinations of elements in water samples collected in 2017 suggested that the water of Chornobyl and Narodychi district did not contain alarming levels of pollution of toxic elements, compared with other cities of Ukraine, Belarus and Tokyo; not only for drinking water but also for hand-washing water. Elemental characteristics of mineral water sometimes showed quite different patterns from other types of water from the same region, suggesting differences in the original water source. For drinking water, purification or disinfection methods, or materials of containers are thought to affect the element contents.

Among alkali metals, a maximum value of Na about 40 mg/L was observed in mineral water bought at Minsk railroad station. A health-based guideline value for Na has not been derived by WHO [1], and this value was much lower than the 200 mg/L (200,000 μg/L) of the average taste threshold for sodium, which is within the water quality standards of tap water in Japan [3] for usability in terms of taste. Potassium concentrations of well water A and C, 36 and 43 mg/L, were much higher than those of other samples. Drinking water quality standard of K has not been fixed by WHO [1]. Potassium is an essential element for humans, however the estimated amounts from drinking water were very small and were not of health concern.

Cesium was measured as a stable isotope <sup>133</sup>Cs by ICP-MS, and was scarcely detected in all samples. Radioactive Cesium <sup>134</sup>Cs and  $^{137}$ Cs were released by the accident [5]; however, we do not discuss short-lived  $^{134}$ Cs (2 years of half-life) here because there are no radiological concerns after more than 30 years. The possibility of the existence of a detectable level of radioactive  $^{137}Cs$  is thought to be extremely low because the counts at  $m/z$  137 matched the expected counts of <sup>137</sup>Ba estimated from <sup>135</sup>Ba. For the determination of <sup>137</sup>Cs by ICP-MS, chemical separation of isobaric interference such as <sup>137</sup>Ba and pre-concentration will be needed [6]. The long-lived (30 years of half-life)  $^{137}$ Cs and  $^{90}$ Sr have been major components of the aquatic ecosystems except during the first several months after the accident in 1986 [5]. Behavior of released radionuclides has been investigated for a long time [5, 7, 8]. The low risk of consumption of  $137$ Cs via the drinking water pathway, in comparison with external and internal (foodstuff) radiation, has been suggested [5,7]. A recent report on 35 years of radiological monitoring of groundwater in the Chernobyl Exclusion Zone indicated that  $^{137}$ Cs and  $^{90}$ Sr concentrations of well water samples taken in 2019 used by residents in the 10 – 30 km zone were less than the WHO drinking water standard, 10 Bq/L [8]. We reported that the tap water of Semipalatinsk, which was contaminated with radionuclides in the past, did not contain toxic elements presenting health problems [2]. Semipalatinsk in East Kazakhstan, is known as the site of 456 nuclear tests of the Soviet Union's nuclear weapons, conducted from 1949 until 1989. The site was closed in 1991.

Of the alkaline earth metals, Ca, Mg and water hardness calculated as CaCO<sub>3</sub> of individual specimens indicated that the three well waters of Chornobyl showed lower values compared with tap and hand-washing water in this area, suggesting differences between components of surface water and ground water. Three well water samples, mineral water B, drinking water B and C, and Japanese water showed low hardness of 100 or less, but most water in Ukraine and Belarus showed hardness of from 120 to 400. The black fertile soil of Ukraine is known to contain rich humus which retain water and nutrients such as Ca and Mg. Health-based guideline value is not proposed for hardness in drinking-water.

WHO criterion has not been indicated for Zn (12 group) because of no health disorder dangers [1]. Drinking water quality standards of Japan [3] and USEPA [4], however, are 1000 and 5000 μg/L, respectively, for acceptability of drinking water regarding color and taste. Figure 1G suggested that well water A in Chornobyl of 2017 contained about 5000 μg/L Zn and that it is not so tasty for drink, but no problem for health.

Aluminium is abundantly present in the Earth's crust or derived from flocculant during water purification process. WHO criterion has not been indicated, but drinking water quality standard of Japan (200  $\mu$ g/L) [3] and USEPS (50-200  $\mu$ g/L) [4] are indicated for reason of coloring. Some samples in Chornobyl, Kyiv and Minsk exceed 50 μg/L.

Cd (group 12), Pb (group 14), As (group 15) and Se (group 16) concentrations of 2017 samples were much lower than those in of the health-based guideline values of WHO criteria, suggesting elemental safety. The result of La (Figure 1I) suggests that

La exists in the ore or soil of this area and that it is partially dissolved in ground water. The provisional guideline value of U concentration of WHO is 30 μg/L [1], considering chemical toxicity. The average concentration of U in Minsk, 0.53 μg/L, and the highest value of 1.55 μg/L were much lower than guideline value and were not a health problem. We reported that U concentrations of piped, shallow well, and river water from near Aral Sea region of Kazakhstan were from 18 to 20 μg/L, but those of deep well were 0.27 μg/L [2]. The source of U in Kazakhstan has been thought to be the uranium mines in central Asian countries.

Three kinds of well water of the Chornobyl exclusion zone (CEZ) area have been used daily for drinking and cooking by the inhabitants living in this area by their own will. The results of testing well water showed some element features different from the other samples of the same area; lower concentrations of Mg, Ca, and Sr. In addition, one or two of three well waters contained higher concentrations of K, Mn, Zn, and La than the other sorts of samples. The shallow well water is groundwater from the top of hard bedrock and is thought to be easily affected by the surrounding environment. Elemental composition of drinking well water in the Transcarpatian region in west Ukraine indicated high Fe contents (53 – 411  $\mu$ g/L) as common characteristics of the surface and underground water of this area [9]. Elemental composition in underground and surface water is affected by geological features and industrial chemicals. Monitoring data of trace elements in tap water of Kryvyi Rih city in the south of Ukraine, which has an industrial complex, indicated seasonal change of Mn, Cu, and Pb contents [10]. Sampling seasons of the water samples of the present study differed depending on the year, and seasonal changes in rainfall, floods and accompanying sediment movement are thought to affect elements in surface and groundwater. Element concentrations and their yearly or seasonal changes are thought to be different in each well.

Extremely high concentration of Pb of 340 μg/L in well water A was observed in 2019, and the source of Pb and the reason for the increase were unknown; however, this contamination was thought to have been a temporal phenomenon because Pb concentration of the next year, 2020, was reduced to 0.3 μg/L again. In well water A, yearly changes in concentrations of Mn and Zn were also large. In the present results, big problems regarding toxic elements were not observed, except Pb in 2019; however, continuous observation is preferable for health protection of inhabitants.

## **Conclusion**

The concentrations of 40 elements in water are shown in this paper. The water samples were collected around the CNPP in Ukraine, Brest, south-west Belarus, Minsk, the capital of Republic of Belarus and Tokyo, Japan. As is well known, the CNPP caused a tremendously large-scale disaster, although it happened more than 30 years ago. There have been various treatments for the water supply. (We did not address this matter.) Various reports on CNPP concerning this accident have been made elsewhere; therefore, we do not address this matter in this paper.

Our objective water samples were ones which general persons nearby can obtain. Our results propose no special indications. Unfortunately, Ukraine and Russia were plunged into war in February 2022. The situation in Ukraine must have changed greatly. This situation may be beyond our imagination. Our water specimens were collected before the outbreak of the war. We expect that our data will be useful in the near future in providing data on common water conditions in a peace time situation in Ukraine.

## **Author contributions**

Shinohara: Organizing data and writing draft, Matsukawa: Analysis and data curation, Kimura: Visit Ukraine and Belarus, sample collection, Yokoyama: Supervision, Chiba: Visit Ukraine and Belarus, sample collection, advise on draft contents.



Region  $\begin{array}{l} \mathrm{Narodychi\;Raton}\\ \mathrm{(n=1)} \end{array}$ Table 2. | Element concentrations in water samples collected in Ukraine, Belarus and Japan in Sep. 2017

Minsk (n=13)

 $K$ yiv<br>(n=6)



 $Tolyy$ okyo

Brest (n=8)

Table 2A

Chernovyl<br>(n=7)

Narodychi Raion









**Figure 1.** | Element concentration in each water sample. (A)Na, (B)K, (C)Mg, (D)Ca, (E)hardness, (F)Mn, (G)Zn, (H)Pb, (I)La, and (J)U.





**Figure 2.** | Time dependent changes in element concentrations of three well waters and tap water of Chornobyl (2017-2020)

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