Toxic Aluminium and Heavy Metals in Groundwater of Middle Russia: Health Risk Assessment

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Abstract: Two approaches are distinguished in modern ecological monitoring. The first one is physicochemical analysis of environmental objects with respect to maximum allowable concentrations (MACs) of chemical substances, which is performed by standards methods in accordance with state regulations. The second approach (biological monitoring) is based on the methodology of biotesting and bio indication. The task of this work is to create biotests for estimation of Al and other metals toxicity in ground water and to compare these results with physicochemical analysis dates. Risk assessment for heavy metals contaminated groundwater was also performed. Risk assessment was performed accordingly EPA US recommendation and gave results about 90 per 100000 citizens for Al and 402 per 100000 for mixture of different heavy metals. For comparison: risk for earth background radiation for Middle Russia is (Individual dose 1 millisivert per year) consist 5 per 100000 people. It was shown that groundwater consist HCO₃⁻ ions (360 mg/l), sometimes Al compounds 0.21-0.65 mg/l (MAC for Al is 0.5 mg/l for Russia). Other groundwater contain Hg – 0.004 mg/l (MAC – 0.0005 mg/l); Cr – 0.072 mg/l (MAC – 0.05 mg/l); As – less than 0.03 mg/l (MAC – 0.05 mg/l). We developed plant biotest for estimation of groundwater quality with barley roots, tradescatia and others. Some biotests parameters correlate with HCO₃⁻, Cl⁻, SO₄²⁻ and metal ions content positively, for another biotest this correlation is strongly negative. The quality of groundwater near Obninsk and in Kaluga Region is very different but hasn’t been changed since the year 1998.

Introduction

Now the traditional ecological monitoring involves not only physical and chemical analysis of environmental objects connected with a system of maximum permissible concentrations (MPC) and classes of danger of chemical substances, maximum permissible doses and levels (MPD and MPL) of ionizing radiation and electromagnetic fields, but also risk assessment which allows qualitative and sometimes quantitative determination of the impact of anthropogenic factors or their combinations to be performed [1].

At present health risk assessment, i.e. assessment of expected consequences as a result of exposure to any hazardous substance or dangerous effect is one of the essential medical-ecological problems.

Contaminated drinking water is one of the expected sources of human health risk. Intensive operation of water intakes and anthropogenic contamination of groundwater are considered to be the reason for this kind of risk. This caused disorder in the natural regime of groundwater and resulted in deterioration of its quality.

The situation observed in Obninsk and other water intakes of Kaluga region may serve an example of these changes. The MPC excess even in one water sample was established from the following parameters: mineral and microcomponent composition (nitrates, nitrites, ammonium, iron, strontium, manganese, fluorine, barium, cadmium, copper), content of toxic substances (benz(a)pyrene, oil-products), radioactive isotopes (tritium, strontium-90) as well as organoleptic (turbidity, color) and bacteriological properties [2].

Increased concentrations of the above elements and compounds in drinking water harmfully affect human health and cause toxic, mutagenic or carcinogenic effects depending on the substance properties. The expected effect of these substances may be illustrated by risk assessment. Therefore the paper seeks to assess the health risk for inhabitants of Obninsk using drinking water.

Health risk assessment in the analysis of environmental quality involves four basic stages [3].

- Hazard identification
- Exposure assessment
- Evaluation of the “dose-response” relationship
- Risk characterization
Identification of Hazardous Ecological Factors in Groundwater

The following objects were chosen for studies:

- Water from springs near the nuclear power enterprise SRC RF Institute of Physics and Power Engineering near Obninsk; these springs are widely popular among citizens.
- Water from three intakes of the centralized water services of Obninsk.
- Information of the State Sanitary Inspection Centre in Obninsk about Human Health.

At the first stage the ecological diagnostics has been performed of water from the chosen springs by biotesting. Water quality was estimated from the following biological parameters: barley germination, biomass and the length of sprouts and barley root parts, the mitotic index and chromosome cell aberrations in the barley root meristem, wheat coleoptile increment. The diagnostics allowed these springs to be divided into three groups.

The first group includes the springs (No.16 as an example) most distant from city and relatively satisfactory. The second group includes the springs (No.2 is typical) located near Obninsk where according to biotesting water practically doesn’t differ from that in the town water pipelines.

Springs with water considered to be the worst both from hygienic and ecological points of view are referred to the third group (e.g. spring No.7). The smallest increment of wheat coleoptiles and a 2 fold excess in a number of aberrant cells relative to the reference level (pipeline cold water) are observed in phytotests (plants) grown in this water. So, it was assumed that water from the third group exhibits mutagenic and, probably, carcinogenic activity.

Only one spring was chosen from each group. The content of chloride-, sulphate-, nitrate- and carbonate-ions, calcium and magnesium cations and pH, mineralization, general rigidity and gamma-radioactivity were determined by physical and chemical methods. Besides, concentrations of aluminium, beryllium, boron, arsenic, mercury, lead, silver, chromium and zinc were assessed. Table 1 shows some of these data.

The studies have shown that the excess was registered for such substances as beryllium, cadmium, mercury and chromium. According to biological and chemical analysis, water from spring No.7 was found to be the worst. In the future when estimating the risk just this spring will be used because of the greatest number of factors capable of causing adverse human impact.

At the second stage the results of chemical analysis of water from three Obninsk water intakes (1990-2003) have been investigated. The analysis has shown that during last several years the excess of iron (up to 1.9 mg/l), the increased rigidity (up to 7.8mg - eqv/l) and fluorine content close to MPC (1.5mg/l) were observed in all three water intakes. Besides, there were variations in mineralization values (from 280mg/l to 500mg/l), pH (from 6.2 to 7.6) and such components as sulphates (from 14 to 136), nitrates (from 0.04 to 6.3), chlorides (from 3 to 35), but their concentration did not exceed MPC.

So, the assessment of adverse factors has shown that heavy metals are the most essential pollutants of spring water. Other pollutants are not analyzed. For pipeline water the worst parameters are associated with iron content, general rigidity as well as with cadmium and chromium. These factors are taken into consideration at the next stages of risk assessment.

Table 1: Chemical composition of some Obninsk springs

| Spring No. | General Rigidity, mg – eqy/l | Concentration, mg/l | Ag  | As  | Al  | Be  | B   | Cd  | Hg  | Pb  | Cr  | Zn  |
|------------|------------------------------|---------------------|-----|-----|-----|-----|-----|-----|-----|-----|-----|-----|
| Maximum Permissible Concentration | 7.0 | 0.05 | 0.05 | 0.5 | 0.0002 | 0.5 | 0.001 | 0.0005 | 0.03 | 0.055 | 5.0 |
| 2          | 5.6 | 0.001 | 0.01 | 0.13 | 0.0006* | 0.010 | 0.019* | 0.004* | 0.019 | 0.072* | 0.003 |
| 7          | 5.7 | 0.001 | 0.01 | 0.11 | 0.0006* | 0.014 | 0.021* | 0.003* | 0.010 | 0.061* | 0.017 |
| 16         | 6.3 | 0.001 | 0.01 | 0.10 | 0.0005* | 0.006 | 0.019* | 0.008* | 0.013 | 0.004 | 0.015 |

* Element excess over MPC.
According to the Environmental Protection Agency (US EPA) technique we should take into consideration the following. Risk is calculated under the condition of this water consumption every day during the whole human lifetime. The water quality standard for calculating risk is also specified for the same period. The average amount of water used every day for drinking is about 3 liters, the mean body weight is 70 kg. So, under these conditions the dose of a chemical substance taken by a person with drinking water every day is:

$$ADDd = \frac{DW \times C}{BW},$$

where, $ADDd$ is the dose taken with drinking water; $BW$ is the body weight, 70 kg; $C$ is the substance content in water, mg/l; $DW$ is the mean volume of water drank every day, 3 liters.

Let us estimate a daily mean dose of the individual’s uptake of contaminants with water. Calculations have been performed using analyzed data on spring (No.7) and pipeline water. Table 4 presents the data obtained.

### Assessment of the "Dose - Response" Relationship for Inhabitants of Obninsk

This stage of activities seeks to represent the quantitative relationship between impact and risk appeared. A linear model suggested by US EPA for risk assessment in our case can be expressed by the following formula:

$$Risk = ADDd \times UR,$$

where, $Risk$ is the risk of adverse health effect estimated as the probability of this effect under given condition; $ADDd$ is the daily substance dose, mg / kg; $UR$ is the risk unit specified as a factor of risk proportion depending on the available concentration (dose). The risk unit $UR$ accepts the true value depending on the impact (carcinogenic, non-carcinogenic) which this substance has and the substance itself.

Table 2 shows the calculated health risk in cases of using spring and pipeline water. Water contains many different components and the risk of a combined impact of contaminants (see Table 4) can be determined from the formula:

$$Risk_{sum} = 1 - (1 - Risk_1) \times (1 - Risk_2) \times \ldots \times (1 - Risk_n),$$

where, $Risk_{sum}$ is the risk of a combined impact of contaminants; $Risk_1$ ... $Risk_n$ is the risk of impact of each isolated contaminant.

As a result of spring water consumption the risk of oncological diseases will come to $3.95 \times 10^{-3}$ and non-oncological diseases to $0.98 \times 10^{-3}$. In terms of per capita (approximately 1000 persons) it means that 4 persons would be in danger of oncological diseases and 1 person of non-oncological diseases.

### Table 2: Calculated carcinogenic and noncarcinogenic risk of groundwater

| Water source   | Substance | C, mg/l | ADDd, mg/kg | UR<sub>cancer</sub>, kg–day/mg | Risk<sub>cancer</sub> | UR<sub>noncancer</sub>, kg–day/mg | Risk<sub>noncancer</sub> |
|---------------|-----------|---------|-------------|-------------------------------|----------------------|----------------------------------|---------------------------|
| SpringNo.7    | Aluminum  | 0.095   | 0.00407     | -                             | 0.1                  | 407×10⁻⁶                          |
|               | Beryllium | 0.0009  | 0.000385    | 4.3                           | 165×10⁻⁶             | 0.002                            | 0.07×10⁻⁶                 |
|               | Boron     | 0.06    | 0.00257     | 0.6                           | 1542×10⁻⁶            | 0.09                             | 231×10⁻⁶                 |
|               | Cadmium   | 0.024   | 0.00103     | 0.38                          | 319×10⁻⁶             | 0.0005                           | 0.52×10⁻⁶                 |
|               | Arsenic   | <0.03   | 0.00128     | 1.5                           | 1920×10⁻⁶            | 0.003                            | 3.8×10⁻⁶                 |
|               | Mercury   | 0.004   | 0.000171    | -                             | 0.0003               | 0.51×10⁻⁶                        |
|               | Lead      | <0.01   | 0.000429    | 0.0085                        | 3.6×10⁻⁶             | 0.0000785                        | 0.3×10⁻⁶                 |
|               | Silver    | <0.001  | 0.000043    | 0.005                         | 0.21×10⁻⁶            | 1.7                              | 73×10⁻⁶                  |
|               | Chromium  | 0.049   | 0.0021      | 0.005                         | 10×10⁻⁶              | 0.1                              | 210×10⁻⁶                 |
|               | Zinc      | 0.004   | 0.000171    | -                             | 0.3                  | 51×10⁻⁶                          |
| Pipe-line Water | Iron    | 0.13    | 0.0056      | -                             | -                    | -                                | -                         |
|               | Cadmium   | 0.0002  | 0.0000086   | 0.38                          | 3.3×10⁻⁶             | 0.0005                           | 4×10⁻⁶                   |
|               | Chromium  | 0.008   | 0.00034     | 0.005                         | 1.7×10⁻⁶             | 0.1                              | 34×10⁻⁶                  |
In case of pipeline water resources the carcinogenic risk of a combined impact amounts to \( 5 \times 10^{-6} \) and non-carcinogenic risk to \( 34 \times 10^{-6} \), that for the inhabitants of Obninsk means 0.54 and 3.7 cases of carcinogenic and noncancerogenic effects, respectively.

**Aluminum in Spring Water - A Source of Risk for Health?**

Until recently it was taken as an axiom that aluminum was harmless for people, animals and plants. In fact, in its content in the Earth's crust (8.8 \%) aluminum is the third after oxygen and silicon and the most widely spread among metals. Thanks to its extremely high reactivity, aluminum quickly forms insoluble compounds and becomes practically safe for plants and animals as it doesn't penetrate into their cells and tissues. Under certain conditions, however, e.g. under the action of acid rains, aluminum can pass to the ionic state and react with biological objects changing them or suppressing their function [4].

Data published in the 1990s have shown that high concentrations of inorganic (free) aluminum were found in the surface and ground water owing to acid rains or any other reasons.

Recent observations show that aluminum content in ground water near Obninsk has increased. It is possibly caused by the above reasons. So, aluminum content in springs in the late 1990s was as follows (table 3).

**Table 3:** Aluminium content in spring water near Obninsk

| Spring No. | Content, mg/l |
|------------|---------------|
| 1          | 0.21          |
| 2          | 0.65          |
| 3          | 0.65          |
| 7          | 0.57          |
| 9          | 0.32          |
| 16         | 0.22          |

The chemical risk will be estimated from these data using the technique proposed (table 4).

**Table 4:** Calculated health risk for drinking water containing aluminum

| Spring No. | C, mg/l | ADDd, mg/kg | UR, kg-day/mg | Risk noncervic |
|------------|---------|-------------|--------------|---------------|
| 1          | 0.21    | 0.0090      | 0.1          | 9.0 \times 10^{-4} |
| 2          | 0.65    | 0.028       | 0.1          | 2.8 \times 10^{-3} |
| 3          | 0.65    | 0.028       | 0.1          | 2.8 \times 10^{-3} |
| 7          | 0.57    | 0.024       | 0.1          | 2.4 \times 10^{-3} |
| 9          | 0.32    | 0.014       | 0.1          | 1.4 \times 10^{-3} |
| 16         | 0.22    | 0.0094      | 0.1          | 9.4 \times 10^{-4} |

The obtained data show that human health risk when drinking spring water with the increased aluminum content ranges from 0.94 to 2.80 cases (per 1 thousand of spring water consumers) of non-carcinogenic effects depending on the number of a spring where water is taken. Though the scientific literature does not cite the unit values to calculate the potential carcinogenic peroral risk for aluminum, the information is available that the increased aluminum content in drinking water may cause chromosome aberrations in barley meristem cells [4]. Therefore further studies of mutagenic and carcinogenic impact of aluminum on living organisms are to be carried out.

Processed data show that water with the increased aluminum content can cause approximately three times as many negative non-carcinogenic effects for human health than other substances contained in spring water and possessing non-carcinogenic effects. At the next stage we tried to associate the physical and chemical parameters of water with the population sickness rate to demonstrate clearly the effect of water quality on human health.

**Ecological-Epidemiological Risk Assessment of the Drinking Water Contamination Impact on the Sickness Rate**

High levels of general rigidity of drinking water, typical of Obninsk can have a direct bearing on human health. Russian and foreign scientists disagree on the impact of this factor on human health. Besides, there are no criteria in literature for quantitative assessment of the expected risk associated with increased drinking water rigidity. Therefore we tried to associate the rigidity parameter with the sickness rate on the basis of ecological and epidemiological risk assessment of drinking water contamination [5].

Harmful environmental impact on human health reveals itself as the increased sickness rate. To study human health we used an ecological approach where temporal variations in the sickness rate were compared with those in the environmental contamination levels. This approach is based on the information from “Report on the cases of diseases registered for patients living in the area of Obninsk medical services». Among a great variety of nozological forms of diseases the approach takes account of those which are most widely spread, adequate to the harmful effect and pollutants; considered are also the informative sanitary parameters available.

In our case we tried to establish the relationship between communication of diseases and drinking water rigidity in the centralized water supply. Epidemiological data on the disease structure and cases for inhabitants of Obninsk served the basis for these studies.

Comparison of the water rigidity and the disease structure allowed certain relationships to be established. So, genito-urinary, stomach ulcer and duodenum diseases, cerebrovascular diseases and diseases of skin and hypodermic tissue are closely connected with this parameter of drinking water. The correlation coefficient for them is 0.70 – 0.94.

Based on the correlation analysis it is possible to conclude that the rigidity value has 4 of 11 important correlation coefficients with the population sickness rate indices in Obninsk. It means that probably each third sickness rate index most probably is associated with drinking water rigidity.
Conclusion

Our data confirm that the risk of diseases in case of spring water consumption is hundreds times higher as compared to pipeline water. Uncontrolled consumption of spring water unfavorable in its composition may be dangerous for human health. According to these studies, pipeline water is more preferable to people, but some risk of carcinogenic and non-carcinogenic effects for inhabitants of Obninsk is also not excluded, however, it is 1000 times lower than the real number of cancer cases in Obninsk.

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