RADIOACTIVITY LEVEL AND CONCENTRATION OF METALS IN WATERS AROUND POWER PLANTS Application of Potential Method for Pollution Assessment

by

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Scientific paper

http://doi.org/10.2298/NTRP1801117S

Human activities may lead to increased levels of naturally occurring radioactive material and heavy metals in waters relative to undisturbed natural environment, consequently to reduced safety of environment. Therefore, evaluation of water quality from coal-fired power plants is essential to both environmental protection and protection of the human health. In this paper the concentrations of metals (K, Sr, and Pb) and corresponding radionuclides (⁴⁰K, ⁹⁰Sr, and ²¹⁰Pb) for 22 water samples in the vicinity of five coal-fired power plants in Serbia were determined. The total metal concentrations were measured using an atomic absorption spectrometer. The activities of radionuclides ⁴⁰K and ²¹⁰Pb were determined by gamma spectrometry using HPGe detector, while the activity of ⁹⁰Sr was determined by radiochemical analytical method and measured using low-level gas proportional counter. The water pollution status was assessed by the prospective method that is defined in this study. The preliminary results indicated that operation of the coal-fired power plants has no significant impact on the surrounding environment with regard to the radiological characteristics and content of metals of the investigated waters that can be used for suitable in future applications.

Key words: metal content, radionuclide, wastewater, pollution

INTRODUCTION

Naturally occurring radioactive materials (NORM) form a part of the natural background radiation to which all humans are exposed. The NORM in the soil and water bodies of an environment are present mostly as progeny of ²³⁸U, ²³⁵U and ²³²Th-series and ⁴⁰K isotope, which are distributed by natural geological and geochemical processes [1]. In addition to the notion of NORM, the concept of technologically enhanced naturally radioactive materials (TENORM) was introduced in mid-seventies because of rapid development of nuclear physics and increasing radioactive material contamination [2]. TENORM is naturally occurring radioactive material whose radionuclide concentrations or potential for human exposure have been increased by human activities much above the levels encountered in the undisturbed natural environment.

One of the main sources of TENORM are coal-fired power plants [3]. Coal combustion in power plants may enhance environmental pollution levels in their vicinity due to fly ash and bottom ash, which are released into the surrounding environment as main solid wastes of coal combustion [4]. This process generates a numerous of pollutants, such as oxides of sulphur, nitrogen and carbon, as well as the toxic and heavy metals and organic particles. Beside this, in the case of coal-fired power plants from this study, the ash mainly is transported to the dump after being suspended in surface water, while all water-surplus is drained back to the river [5]. Radioactivity present in coal and ash, like in most materials found in nature is mainly due to the presence of radioactive elements [6]. The primary environmental concern associated with radionuclides coming from a field disposal site is potential groundwater contamination [5].

Besides presence of the natural radioactivity, a certain amount of artificial radionuclides is ejected into the atmosphere. The level of contamination of radionuclides

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The pollution of water systems affects humans and the natural environment worldwide. The sustainable use of water is a highly important subject worldwide and the necessity of reducing water pollution and waste generation is of prime importance for the future of human kind [8, 9]. Among the wide diversity of pollutants affecting water resources, heavy metals have particular role considering their strong toxicity. Occurrence of metals in surface waters can be due to natural sources (precipitation and geochemical sources, *i. e.* dissolution of naturally occurring minerals containing trace elements in the soil zone). Anthropogenic sources such as mining, fuels, smelting of ores and improper disposal of industrial and municipal wastes, significantly contribute to elevated levels of metals in waters [10, 11].

Due to the fact that coal-fired power plants are located in around the populated areas and the work process of coal-fired power plants causes significant environmental pollution, the impact of the environment on the residential population can be significant. It is necessary to control coal-fired power plant's environment. It is required to systematically monitor the changes, as well as the possibility to assessing the impact of power plants in relation to possible increase of the level of background radiation in the environment. Due to the above mentioned reasons, this paper deals with the analysis of different kind of waters from coal-fired power plants and their surroundings. The analysis results are important considering that the Serbian population is dominantly supplied with water from some of the rivers endangered by operations of coal-fired power plants. The content of Sr, K, and Pb in waters surrounding of the coal-fired power plants was determined, with special attention to their corresponding radioactive isotopes: 90Sr (one of the most biologically hazardous artificial radionuclides and as the presenter of "nuclear" pollution, ⁴⁰K (due to terrestrial origin, as well as, its good solubility in water) and ²¹⁰Pb (as natural gamma emitter with an origin in atmospheric deposition; radon gas, as well as soil suspension), in order to identify the origin and estimation of key pollution sources which affect these waters [12, 13]. Besides that, the objective is to provide insight into the current condition of water systems and to propose a potential method for pollution assessment.

MATERIALS AND METHODS

Sampling and sample preparation

The sampling of 22 water samples (overflow of ash deposit, drain and river waters, as well as one sample of drinking water) was performed during October 2014

at different locations from the following coal-fired power plants and their environs in the Republic of Serbia: "Nikola Tesla A and B" (TENT A and TENT B, Obrenovac), "Morava" (TE MORAVA, Svilajnac), "Kolubara" (TE KOLUBARA, Veliki Crljeni) and "Kostolac" (TE KOSTOLAC, Kostolac). The map of sampling sites is shown in fig. 1. The volume of each sample was about 20 L. About a liter of each sample was taken to determine the total metal content, while the rest was evaporated to approximately 200 mL under infrared lamps and then filtered for the radionuclide content analysis, apropos for the gamma spectrometric measurement and radiochemical analytical procedure of 90Sr determination. To determine total dissolved solids (TDS), samples were refrigerated at 4 °C until the analysis and during analysis, filtered through Reeve Angel type 934-AH glass fiber filter discs without an organic binder.

Methods of analysis

All water samples were analyzed for pH, electrical conductivity (EC) and TDS dried at 180 °C. For pH measurement pH meter pH/°C Tester pHep4 (Hanna Instruments, USA) was used. Calibration of the instrument was carried out by 4, 7, and 10 pH standard solutions. The EC measurements were done by conductometer CON 400 (Oakton Instruments, USA) with a cell constant of 1.0. The measuring range 0-1999 S was calibrated at 1411 S using standard 0.01 M KCl solution at 25 °C. TDS was measured following EPA Method 160.1 [14].

The total metal concentrations were determined by Atomic absorption spectrometry (AAS). Before the analysis, the samples were filtered through membrane filter and then acidified to pH 1 with 65 % HNO₃. Also, the preparation of the samples for determination of K involved the addition of certain concentration of Cs modifier, while for determination of Pb, certain concentration of magnesium nitrate modifier was added. The stock solutions were purchased from MBH Analytical LTD (Strontium Standard), Fluka Analytical Sigma Aldrich (Potassium Standard) and Baker Analysed (Lead Standard). Multistandard for curve control was purchased from Merck. The analysis of the total metal concentrations was performed using Atomic absorption spectrometers SensAA 2G and SensAA(GBC scientific equipment) in which the inert nitrogen gas and air-acetylene flame were used, sequentially. The concentrations of Sr and Pb were determined by Graphite furnace technique [15], while K by Flame technique. The type of the lamp for determining K and Pb was S & J Juniper & Co (United Kingdom); the used wavelengths were 769.9 and 330.2, sequentially. PHOTRON lamp (Australia) was used for the determination of total Sr at 460.4 nm wavelength.

The determination of ⁹⁰Sr activity concentration in water samples was performed by validated



Figure 1. The map of sampling sites

radiochemical analytical method and the applied method and the calculations have been described in a previous paper [16]. The ⁹⁰Sr activity in the samples was determined by measurement of the activity of 90Y using α/β low-level proportional counter (ESM Eberline Instruments GmbH, Germany) with system counting efficiency of 35 % for determination of the activity of beta emitters. The beta efficiency of gas proportional counter was checked with ⁹⁰Sr standard source. The counting time was 3600 s in five series of measurements. The gamma ray spectrometric measurements of ⁴⁰K and ²¹⁰Pb radionuclides were performed using HPGe detectors (Canberra Industries, USA) with 18 and 20 % counting efficiency. The GE-NIE 2000 program (Canberra Industries, USA) was used for the spectra analysis. The activities of ⁴⁰K and ²¹⁰Pb were determined from their energy lines of 1460 and 46 keV, respectively. The details about the calibration of detectors, the spectral analysis and the calculations have been described in an earlier paper [17]. The measurement time was 60 000 s.

RESULTS AND DISCUSSION

The results of pH determination, electrical conductivity and TDS in water samples are listed in tab. 1. It is evident from tab.1 that all water samples are alkaline (pH 7.7 to 12.5), except for a single groundwater sample (pH 5.4). Also in tab. 1, it can be seen that EC values ranged from 423 to 3030 Scm⁻¹. The two water samples from TE MORAVA are extremely alkaline (pH 12.4 and 12.5) and these samples have the highest EC values. The sample of groundwater from TE KOSTOLAC is acidic and also has high value for electrical conductivity (2280 Scm⁻¹). The pH and electrical conductivity values strongly depend on the dissolved substances in the water [18]. Due to higher limiting H⁺ ion conductivity than OH⁻ ion conductivity acidic solution is generally better conductor [19]. In our case, water with higher pH value has large EC as well. The explanation for such behavior should be in the greater concentration of dissolved alkali salts and these samples probably have abundant different metal

Power plant	Location	pН	EC [μ Scm ⁻¹]	TDS $[mgL^{-1}]$
TENT A	Landfill overflow water	8.3	851	568
	Drain water at well	8.4	1343	940
	Drain water between wells	8.4	1407	985
	Sava river upstream	8.5	439	240
	Sava river downstream	8.5	423	232
	Landfill overflow water	8.3	659	447
TENT D	Drain water	8.0	1271	890
IENI B	Sava river upstream	8.9	442	251
	Sava river downstream	8.3	460	252
	Landfill overflow water	12.4	3030	2057
	Drain water	12.5	2710	1951
IE MORAVA	Morava river upstream	8.7	448	262
	Morava river downstream	9.0	462	265
	Landfill overflow water	7.9	1405	1012
	Drain water	8.4	1236	853
TE KOLUBARA	Turija river upstream	8.3	619	379
	Kolubara river downstream	8.3	630	371
	Drinking water	7.7	798	507
TE KOSTOLAC	Landfill overflow water	8.0	598	407
	Landfill drain water	7.9	572	394
	Drain water (Drmno)	8.1	685	413
	Drain groundwater (Cirikovac)	5.4	2280	1642
Recommended values [20, 21]		6.5-9.5	<2500	

Table 1. The values of pH, electrical conductivity and total dissolved solids in investigated water samples

ions (such as Na⁺, Li⁺, K⁺, Mg²⁺, Ca²⁺). This is in accordance with similar studies [5,18]. Other contaminants, such as ammonium ion should not be excluded. According to regulation on quality and other requirements for natural mineral water, spring water and bottled drinking water [20, 21], the obtained results are pointing out that three pH values are outside of recommended range, while for EC only two values are slightly higher than recommended value and there is no indication of the coal-fired power plants' influence on the water pollution.

The results of analysis on total metal concentrations in water samples are presented in tab. 2. The concentrations of micro element Sr ranged from 0.1 to 3.1 mgL⁻¹. Similar levels of total Sr content have been found in study [22] in Danish surface waters and the values ranged between 0.06 and 1.05 mgL⁻¹. However, for this component in water samples, there is no regulation on maximum allowed concentration, but the US EPA (United States Environmental Protection Agency) has set a health reference level for strontium [23, 24]. In 2014 the health reference level of 1.5 mgL^{-1} was establish, but did not finally adopted. This value was calculated using the reference dose of 0.3 mgkg⁻¹d⁻¹ [23]. Currently EPA recommends that level of stable strontium in drinking water should not be more than 4 mgL⁻¹ [24]. The concentrations of micro element Pb are below 0.005 mgL^{-1} , *i. e.*, the minimum detectable concentration (MDC), except for two samples of surface water and one sample of landfill drain water. The recommended value of Pb content is 0.01 mgL^{-1} [20, 21, 25-27]. This criterion is satisfied

by all analyzed water samples, except for one. The content of macro element K varied from 1.5 to 30.8 mgL⁻¹. The highest concentration for K is obtained in the groundwater sample from TE KOSTOLAC. It is in good agreement with above mentioned assumption that the large amount of dissolved minerals in groundwater increase the conductivity and the K is one of main soil ingredients. In Spanish study [2], the concentration of potassium in waste water samples ranged from 5.5 to 46.4 mgL⁻¹, which coincides with the our results. If the K content compares with regulation [25], then 73 % of these waters would meet the criteria as safe.

The activity concentrations of the artificial radioactive isotope 90 Sr, as well as the natural radioactive isotopes 40 K and 210 Pb in investigated water samples are shown in tab. 3. The activity concentrations of 90 Sr ranged between 0.003 and 0.041 BqL⁻¹. The activity concentrations of 90 Sr in water samples analyzed in this study are less than recommended value of 4.9 BqL⁻¹ in drinking water [28]. The radionuclide of 40 K is found in these waters and the activity concentrations were in the range 0.04 to 0.79 BqL⁻¹. The activity concentrations of 210 Pb are below MDC, except in two samples (tab. 3). The analyzed sample of drinking water is in accordance with recommended value of 0.2 BqL⁻¹ [28, 29].

The linear correlations between total metal concentrations, radionuclides and pH, as well as electrical conductivity are determined. Medium positive correlation is obtained between total Sr concentration and electrical conductivity with Pearson correlation coef-

Power plant	Location	$Sr [mgL^{-1}]$	K $[mgL^{-1}]$	Pb $[mgL^{-1}]$
TENT A	Landfill overflow water	0.8	5.5	MDC
	Drain water at well	1.1	12.9	MDC
	Drain water between wells	1.3	13.3	MDC
	Sava river upstream	0.2	1.6	MDC
	Sava river downstream	0.1	3.0	MDC
	Landfill overflow water	0.5	5.2	MDC
TENT D	Drain water	1.2	17.8	MDC
IENI B	Sava river upstream	0.2	1.5	0.007
	Sava river downstream	0.1	1.8	0.009
	Landfill overflow water	1.0	14.6	MDC
	Drain water	1.1	30.8	MDC
TE MORAVA	Morava river upstream	0.2	3.6	MDC
	Morava river downstream	0.1	4.0	MDC
	Landfill overflow water	0.7	11.0	MDC
	Drain water	1.1	3.9	MDC
TE KOLUBARA	Turija river upstream	0.2	3.5	MDC
	Kolubara river downstream	0.2	3.6	MDC
	Drinking water	0.1	3.0	MDC
TE KOSTOLAC	Landfill overflow water	0.4	1.8	MDC
	Landfill drain water	0.3	3.1	0.015
	Drain water (Drmno)	0.2	2.9	MDC
	Drain groundwater (Cirikovac)	3.1	30.2	MDC
Maximum allowed concentration [20, 21, 27]		_	12	0.01

 Table 2. The total metal concentrations in water samples

Table 3. The activity concentrations of investigated radionuclides in water samples

Power plant	Location	90 Sr [BqL ⁻¹]	40 K [BqL ⁻¹]	²¹⁰ Pb [BqL ⁻¹]
TENT A	Landfill overflow water	0.026 0.003	0.18 0.03	<0.1
	Drain water at well	0.027 0.004	0.25 0.03	< 0.07
	Drain water between wells	0.036 0.004	0.44 0.04	< 0.07
	Sava river upstream	0.032 0.004	0.08 0.02	< 0.09
	Sava river downstream	0.015 0.003	0.13 0.04	< 0.04
	Landfill overflow water	0.028 0.003	0.13 0.02	< 0.07
	Drain water	0.041 0.004	0.41 0.04	< 0.07
IENI B	Sava river upstream	0.014 0.002	0.04 0.01	0.18 0.04
	Sava river downstream	0.007 0.002	0.10 0.03	0.23 0.06
	Landfill overflow water	0.024 0.004	0.56 0.05	< 0.09
TEMODAVA	Drain water	0.027 0.004	0.68 0.06	< 0.09
TE MORAVA	Morava river upstream	0.016 0.003	0.12 0.03	< 0.1
	Morava river downstream	0.008 0.002	0.13 0.03	< 0.06
TE KOLUBARA	Landfill overflow water	0.022 0.003	0.30 0.03	< 0.06
	Drain water	0.017 0.003	0.14 0.03	< 0.1
	Turija river upstream	0.014 0.003	0.16 0.03	<0.1
	Kolubara river downstream	0.008 0.002	0.14 0.03	<0.1
	Drinking water	0.006 0.002	0.11 0.02	< 0.06
TE KOSTOLAC	Landfill overflow water	0.005 0.001	0.24 0.03	< 0.07
	Landfill drain water	0.004 0.001	0.14 0.03	<0.1
	Drain water (Drmno)	0.003 0.001	0.21 0.03	< 0.07
	Drain groundwater (Cirikovac)	0.032 0.004	0.79 0.07	< 0.08
Derived activity concentration [20, 21]		4.9	_	0.2

ficient (*r*) of 0.713. Weak positive correlation is obtained between ⁹⁰Sr activity concentration and electrical conductivity (r = 0.472). Between the total K content, as well as ⁴⁰K activity concentration and elec-

trical conductivity, the obtained dependencies are strongly positive, with the correlation coefficients of 0.854 and 0.898, respectively. The obtained results confirmed that K^+ ion has important contribution to



Figure 2. Dependence of 90 Sr activity concentration on total Sr concentration (a); Dependence of 40 K activity concentration on total K concentration for investigated water samples (b)

electrical conductivity. The investigated metals and radionuclides have not shown correlation with pH. Among others, the correlations between ⁹⁰Sr activity concentration and total Sr concentration, ⁴⁰K activity concentration and total K concentration are determined (fig. 2). Medium positive correlation (r=0.680) is found between ⁹⁰Sr activity concentration and total Sr concentration for investigated water samples. However, strong positive correlation is found between the ⁴⁰K activity concentration and total K concentration in investigated water samples with the correlation coefficient of 0.947.

In addition, the pollution assessment, from investigated coal-fired power plants is done. In the absence of wastewaters and river water regulations, as well as, time dependent measurements (considering that presented work is the first research of this kind in Serbia), we have defined a method for verifying pollution from power plants near rivers. The obtained results strongly depend not only on chemical composition of investigated water, but also of the soils, the type of vegetation, soil solution, geology, atmosphere, etc. near sampling sites [30]. According to above mentioned, for pollution assessment, it seems reasonable to compare only river upstream and downstream from power plant, considering that power plants use river water for various processes. For that purpose, the [^{*}M]/[M] ratio is defined in this study. The [^{*}M] is the

concentration of investigated radioactive nuclide and [M] is total metal concentration obtained with AAS. Our main idea was that [*M]/[M] ratio of the river upstream and river downstream remains the same value if the contamination of river water has not occurred by power plant. It could be taken for ⁴⁰K and ²¹⁰Pb as natural radionuclides, but also for ⁹⁰Sr as artificial one. Since in context of the content of 90Sr cannot be consider as TENORM source [2], the ratio for [*Sr]/[Sr] river upstream and downstream from power plant should constant or lower due to Sr present in coal combustion residuals and waste waters (without ⁹⁰Sr). Therefore, the measuring Sr ratio in the upstream and downstream river water's sample can be used as a probe (or test sample) for proposed method. If the basis of established assessment is well, then the [*Sr]/[Sr] ratio should be constant for particular river downstream and upstream water, due to power plants are not the potential source of Sr contamination. However, it is well-known that the disposal of coal ash, incinerator ash and industrial wastes may increase the concentration of strontium in soil [24]. A part of stable and radioactive Sr in soli may dissolve in water, so it is likely to move deeper into the ground and enter groundwater. Therefore, it is very important to exclude river creek and groundwater as potential source of contamination and giving false results for power plant pollution. This should be avoided by the careful selection of upstream and downstream sampling point/place.

The obtained results of the assessment of coal-fired power plants pollution are shown in tab. 4. Considering that the obtained results for activity are in BqL^{-1} , concentration of metal radionuclide is [*M] = $= k_M A_M$, where A_M is the activity concentration (in BqL^{-1}) of investigated radionuclide and k_M is constant of proportionality (same for particular metal). Therefore for comparison, the A_M/[M] upstream and downstream were taken. This correlation is given assuming that, due to small value for total metal concentration (meaning absence of saturation), all obtained A_M values belong to the linear part of the calibration curve $[^*M] = f(A_M)$. The obtained upstream and downstream $A_M/[M]$ rations for Sr, K and Pb are presented in tab. 4. If there is no pollution of the investigated metals and appropriate radionuclides, this ratio should not change significantly. It is obvious that contamination of river water from coal-fired power plants has not occurred,

Table 4. The assessment of coal-fired power plants pollution: ratio of the activity concentration of radionuclide $[BqL^{-1}]$ and total concentration of the corresponding metal $[mgL^{-1}]$

Power plant	Location	A _{Sr-90} /[Sr]	A _{K-40} /[K]	A _{Pb-210} /[Pb]
TENT A	Sava river upstream	0.160	0.050	_
	Sava river downstream	0.150	0.048	-
TENT B	Sava river upstream	0.070	0.027	25.7
	Sava river downstream	0.070	0.033	25.6
TE MORAVA	Morava river upstream	0.080	0.033	-
	Morava river downstream	0.080	0.032	_

as same values for ⁹⁰Sr/Sr, ⁴⁰K/K and ²¹⁰Pb/Pb upstream and downstream from the coal-fired power plants are observed for particular river water.

The proposed assessment has shown good potentiality for application, but the data are presented only for three samples and further investigation, including larger number of samples, as well as samples from each site at different times is needed. However, the orientation criteria for contamination should be done considering the maximum allowed values of activity concentrations for drinking water and total metal concentration obtained in this work. Thus, if mentioned ratios for downstream river water are higher significantly than values of ratio for the upstream river, then we can consider the potential pollution from power plants.

CONCLUSION

The survey of radionuclide concentrations (⁹⁰Sr, ⁴⁰K, ²¹⁰Pb) and total metal concentrations (Sr, K, Pb) assessment in waters from around the coal-fired power plants in Serbia has been carried out. According to conducted study focusing on analysis of content of metals and the corresponding radionuclides and control of environmental waters from surrounding of the coal-fired power plants, it can be concluded that the obtained results do not show the increased pollution of the environment due to operation the power plants. The prospective new method for verifying pollution of waters from coal-fired power plants is described. Presented method has shown the good potentially for application, but further investigation is needed.

ACKNOWLEDGEMENT

The authors would like to thank to the Ministry of Education, Science and Technological Development of the Republic of Serbia for material support (Grants no. III 43009, III 45014 and 172015).

AUTHORS' CONTRIBUTIONS

The manuscript was written by N. B. Sarap and M. C. Pagnacco. Experiments were done by all authors. The results were analyzed and discussed by N. B. Sarap and M. C. Pagnacco. The tables were prepared by N. B. Sarap. The figures were prepared N. B. Sarap and M. C. Pagnacco.

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Received on August 22, 2107 Accepted on March 8, 2018

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НИВО РАДИОАКТИВНОСТИ И КОНЦЕНТРАЦИЈА МЕТАЛА У ВОДАМА У ОКОЛИНИ ТЕРМОЕЛЕКТРАНА Примена потенцијалне методе за процену загађивања

Људска активност може довести до повећања нивоа природног радиоактивног материјала и тешких метала у водама у односу на природно окружење у ненарушеном стању, чиме се повећава ризик угрожености животне средине. Због тога је процена квалитета вода из окружења термоелектрана од суштинског значаја, како за заштиту животне средине, тако и за здравље људи. У овом раду утврђене су концентрације метала (K, Sr и Pb) и одговарајућих радионуклида (40 K, 90 Sr и 210 Pb) за 22 узорка воде у близини пет термоелектрана у Србији. Концентрације метала су измерене помоћу атомског апсорпционог спектрометра. Концентрација активности радионуклида 40 K и 210 Pb одређене су методом гама спектрометрије помоћу германијумског детектора високе чистоће, док је концентрација активности 90 Sr одређена радиохемијском аналитичком методом и мерена нискофонским гасним пропорционалним бројачем. Ниво загађености вода је процењен предложеном методом, дефинисаном у овој студији. Прелиминарни резултати показали су да рад термоелектрана нема значајнијег утицаја на животну средину у погледу радиолошких карактеристика и садржаја метала испитаних вода, што их може учинити погодним за будуће примене. Имајући у виду важност очувања вода, приказани резултати могу бити од помоћи у предикцији и елиминисању фактора који доводе до ризика од загађења.

Кључне речи: садржај мешала, радионуклид, ошиадна вода, загађење