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ARTICLE

Spatial Investigation of Nilüfer Stream Arsenic Pollution in Previous and Post COVID-19 Pandemic and Evaluation of Health Risks for Adult People

Aslıhan Katip*

Faculty of Engineering, Department of Environmental Engineering, Bursa Uludag University, Bursa, 16059, Turkey

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ABSTRACT

This study was carried out in Nilüfer Stream in Bursa City, where intensive industrial, agricultural and mining activities are existed. The temporal and spatial variation of arsenic was evaluated by examining its concentrations between March 2015 and December 2021. Values between March 2015 and December 2019 were evaluated as pre-pandemic, and values between March 2020 and December 2021 were evaluated as post-pandemic. The results were compared with national and international standards and the chronic and cancer risks were calculated for adults. When the 7-year general averages were examined, it was seen that the highest concentration was 0.0256 mg/L at the 8th Station, and the lowest concentration was 0.0182 mg/L at the 1st Station. The reason why the highest value is at the 8th station was that the wastewater of Nilüfer and Bursa Organized Industrial Zones was discharged to Bursa West Wastewater Treatment Plant before this station. After the pandemic the raises in concentrations were observed at all stations, except for the 3rd Station. This shows that the pollution load had increased in general during the pandemic. However, it was estimated that there was a decrease in the pollution load of the industrial wastewater coming to the 3rd Station, which was located after the Eastern Wastewater Treatment Plant of the City. It was observed that all stations examined were higher than drinking water standards and lower than irrigation water standards according to WHO and Turkish National Standards. All measuring stations were greater than 1 of the hazard quotient (HQ) values. In terms of human consumption risk, all stations had a chronic and carcinogenic risk according to the values before and after the pandemic. After the pandemic conditions, the HQ order of the stations was 8>10>7>9>2>6>4>1>3. In general, post-pandemic HQ values had generally increased and the risk of cancer had increased.

*Corresponding Author:
Aslıhan Katip,
Faculty of Engineering, Department of Environmental Engineering, Bursa Uludag University, Bursa, 16059, Turkey;
Email: aballi@uludag.edu.tr

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1. Introduction

Arsenic occurs naturally in the earth's crust and is an element classified as a semi-metal or metalloid. Arsenic could enter waters from pesticides and phosphate fertilizers, mine drainage, oxidation of arsenic-containing sulfide minerals, reduction of arsenic-bearing iron and manganese oxides, discharges of geothermal waters and power plants, improper production, use and disposal of arsenic-containing products [1,2]. Industrial productions containing arsenic include wood, timber preservation, cosmetics, paint enterprises, pharmaceutical industry, herbicide industry, semiconductor material production, leather, glass production, medical uses, paper and pulp production, and cement enterprises. In addition, copper, nickel, gold mining and ore disposal operations, agricultural practices, use of fossil fuels, landfill leachate are among the anthropogenic sources of arsenic [3,4].

In the report titled "Human Development Report 2006 on the Verge of Scarcity: Power, Poverty and the Global Water Crisis" prepared by the United Nations Development Program, Turkey had been shown among the countries with the possibility of arsenic contamination [3]. High levels of arsenic had also been detected in Bursa and its environs, as well as in Balıkesir and Uşak [5,6]. The Nilüfer Stream, which passes through Bursa, where the textile, automotive, metal and chemical industries were intense, agricultural production and mining was carried out, and which was the 4th largest city of Turkey, is under intense pressure in terms of metal pollution [7-9]. There was Emet and Orhaneli Stream within the borders of Bursa City. In the Uluabat Lake Basin formed by the Emet and Orhaeli Streams, there were Keles Lignites Enterprise, Tunçbilek Coal Enterprise, Tunçbilek Thermal Power Plant and Emet Colemanite enterprises on the Emet Stream. It was stated in the literature that the colemanites of the region contain arsenic in the form of orpiment and realgar [10]. It was known that mineralized coal types were rich in toxic trace elements such as arsenic, mercury, antimony, and thallium. Arsenic was mixed into the waters in Uluabat Lake and its Basin, where mining was carried out intensively [10]. The 5 wastewater treatment plants located in the Nilüfer Stream Basin discharged the treated industrial and domestic wastewater to the Stream [8]. It was thought that the use of herbicides containing arsenic compounds in Bursa, which was an important agricultural center in Turkey, causes soil and water pollution [11].

Within the scope of this study, the variation of As pollution between the years 2015-2021 at the measurement stations on the side and main branches of the Nilüfer Stream was investigated. Also, it was aimed to observe the effects of the COVID-19 pandemic on As pollution and to examine the concentration trends in the last 7 years. This study was scientifically original because it was shown the effect of the COVID-19 pandemic on As pollution.

2. Materials and Methods

2.1 Study Area

Nilüfer stream which arises from the border of Bursa city served as an important water supply for the city and reaches the sea after a long flow in the basin. The Nilüfer stream had been exploited to support agriculture and the public water supply. The major sources of pollutants of Nilüfer stream come from point sources which were mainly composed of treated wastewater discharges from organized industrial districts and municipal sewage treatment plants. There were 5 large wastewater treatment plants in the Nilüfer Stream Basin. Two of them (Demirtaş and Bursa Organized Industrial Zones Wastewater Treatment Plant) had a completely industrial wastewater characterization, while the Eastern, Western and Green Environment Wastewater Treatment Plants had domestic and industrial characterization. Also, non-point sources were mainly composed of surface run-off from agricultural areas [7].

In Bursa City, apart from textile, automotive, metal, and chemical industry, wastes arising from the processing of many marble quarries, tungsten, lignite, boron, magnesite, zinc, asbestos, chromium, and olivine ores were found in the basin. In addition, there was 1 coal-fired thermal power plant in the province [12].

Samples were taken from 10 different points, upstream and downstream of the Nilüfer Stream, before and after the wastewater treatment plants. The locations of the measuring stations were given in Table 1 and Figure 1.

Table 1. Locations of the measuring stations

Station No	Location
1	On Nilüfer Stream, Gümüştepe Locality
2	On Deliçay Creek, Before Discharge of East wastewater treatment plant
3	On Deliçay Creek, After Discharge of East wastewater treatment plant
4	On Nilüfer Stream, After Deliçay Creek Mixture
5	İsmetiye Stream
6	On Nilüfer Stream, After Dosab Wastewater Treatment Plant Discharged
7	On Ayvalı Creek Before West Wastewater Treatment Plant Discarharge
8	On Ayvalı Creek, After West Wastewater Treatment Plant Discharge
9	On Hasanağa Creek
10	On Nilüfer Stream, After Hasanağa Creek Mixture

2.2 Laboratory Analyses

Water samples were collected from midstream at a depth of 15 cm ~ 20 cm in 1000 mL polyethylene bottles, which had previously been cleaned by soaking in 10% nitric acid and rinsed with distilled water. At the sampling site, the bottles were rinsed twice with the water to be sampled prior to filling. The water samples were acidified on site to a pH less than 2 with 5 mL of analytical grade concentrated HNO_3. After collection the samples were placed in coolers with ice bags while being transported to the laboratory and kept at about 4 °C until being analyzed [8,13]. Grab samples were collected in dry weather conditions from the 10 measuring stations seasonaly (March, June, September, and December) between 2015 and 2021.

Water samples were implemented digesting process via a CEM MARS-5 model microwave instrument. A 40 mL sample was placed into the cell and then 6 mL of HNO_3 (65% analytical grade) and 4 mL of HCl (37% analytical grade) were added to the cell. 180 psi pressure and 160 °C temperature was applied for 20 minutes to the cell. After cooling 30 minutes to room temperature samples transferred into a 100 mL flask. The digested samples were filled with distilled water to the 100 mL mark, and used in the ICP-AES (Vista MPX, Varian) analysis. The standard calibration solutions were prepared at concentrations of 0.05 mg/L, 0.1 mg/L, 0.25 mg/L, 0.5 mg/L, and 1 mg/L. For higher than 1 mg/L of samples, calibration solution concentrations were prepared at 1 mg/L, 2 mg/L, 5 mg/L, and 10 mg/L. The blanks were done by concentrated 5% HNO_3 into ultrapure water. Quality control was performed with certified liquid samples (multi-elements standard, catalogue number 900-Q30-002, lot number SC0019251, SCP Science, Lasalle, Quebec) to provide the accuracy of the measurements. Quantification limit was 5 μg/L for As. Certified liquid samples were used to check the analytical accuracy, which ranged between 1% and 10%. All reagents used were of analytical grade or better [13].

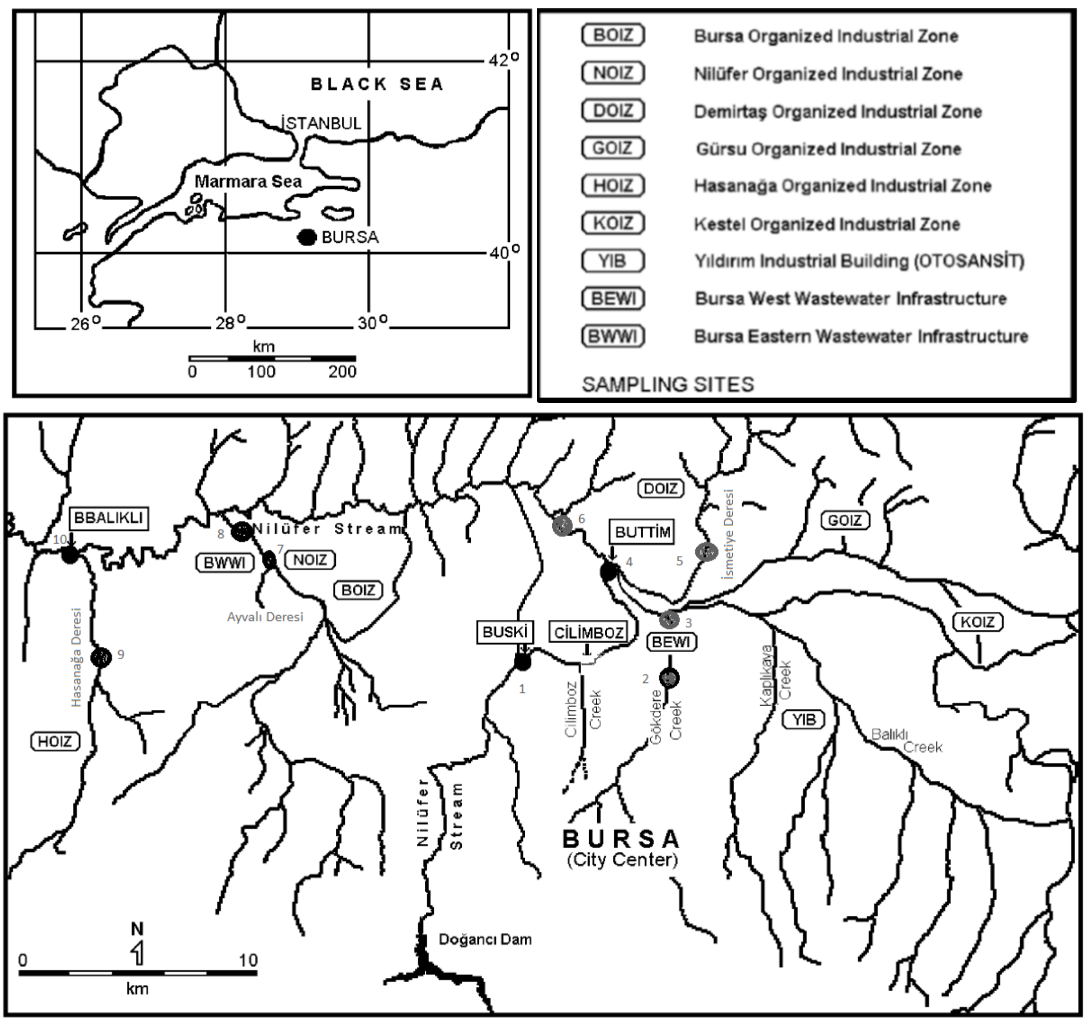

Figure 1. Location of Measuring Stations (Adapted from Güleryüz et al., 2008) [14].

2.3 Evaluation of the Health Risk of Arsenic

Arsenic enters into human body by the way of nutrition, dermal contact and inhalation [15]. The average daily dose (ADD) through potable water intake was estimated according to the following equation [16,17].

$$ADD = C \times IR \times ED \times EF / (BW \times AT) \quad (1)$$

In this equation ADD was the average daily dose during the exposure (mg/kg-day) and C represented the arsenic concentration in water (μg/L), IR was water consumption rate (2 liters for adults and one for children's), ED was duration of vulnerability (70 years for adults and 10 years for children), EF was exposure frequency (365 days' years 1), BW was body weight (72 kg for adults and 32.7 kg for children), and AT was average life time (25,550 days for adults and 3650 days for children) [17,18].

In this study, chronic and carcinogenic risk situation were evaluated. The HQ could be estimated by the following equation [16].

$$HQ = ADD / RfD \quad (2)$$

In this equation, RfD- the toxicity reference dose was 0.0003 mg/kg.day [19]. If the HQ values were >1 the human health risk was exist [17,18,20]. The equation of cancer risk (CR) was as below:

$$CR = ADD \times CSF \quad (3)$$

The cancer slope factor (CSF) of EPA for As is 1.5 mg/kg.day [19]. The CR value greater than one in million (10^{-6}) was generally considered significant by USEPA. All fixed coefficients and reference values from the literature in the calculations were used in μg/L.

3. Results and Discussion

3.1 Spatial Evaluation of Variation of Concentrations

To examine the temporal and spatial variation of arsenic, its concentrations between March 2015 and December 2021 were examined. Values between March 2015 and December 2019 were evaluated as pre-pandemic, and values between March 2020 and December 2021 were evaluated as post-pandemic.

When the values at the measurement stations before and after the COVID-19 pandemic were examined, it was seen that the concentrations at all stations increased, except for the 3rd Station. When the values before the pandemic were examined, it was seen that the highest concentration was 0.213 mg/L at the 10th Station and the lowest concentration was 0.0178 mg/L at the 1st Station. When the post-pandemic concentrations were examined, the highest concentration was again observed at the 10th Station (0.0358 mg/L), and the lowest concentrations were observed at the 3rd (0.0189 mg/L) and 1st and (0.0193 mg/L) Stations. The mean and standard deviation values of As concentrations before and after the pandemic at the measurement stations were given in Table 2.

Table 2. The Mean and standard deviations of pre-pandemic and post pandemic As concentrations

Stations	Pre-Pandemic		Post-Pandemic	
	Mean	Std	Mean	Std
1	0.0178	0.0068	0.0193	0.0053
2	0.0206	0.0056	0.0218	0.0049
3	0.0194	0.0067	0.0189	0.0048
4	0.0195	0.0056	0.0201	0.0047
5	Not Enough Measurements		Not Enough Measurements	
6	0.0192	0.0059	0.0206	0.0046
7	0.0208	0.0063	0.0318	0.0272
8	0.0200	0.0074	0.0358	0.0373
9	0.0196	0.0074	0.0223	0.0060
10	0.0213	0.0066	0.0358	0.0314

It was thought that the reason for the decrease in the concentrations at the 3rd station after the pandemic was the decrease in the pollution load coming to the Eastern Wastewater Treatment Plant because of the decrease in the production in the industry during the pandemic. The general and seasonal averages of all values before and after the pandemic were examined. Accordingly, when the 7-year general averages are examined, it was seen that the highest concentration was 0.0256 mg/L at the 8th Station, and the lowest concentration is 0.0182 mg/L at the 1st Station. When the 7-year spring, summer and winter averages were examined, it was observed that all the maximum values were 0.0221 mg/L, 0.0231 mg/L and 0.0337 mg/L at the 7th Station, respectively. The maximum autumn average was determined as 0.0309 mg/L at the 8th Station. The minimum values in the spring, autumn and winter seasons were determined as 0.0187 mg/L, 0.0181 mg/L and 0.0170 mg/L at Station 1, respectively. The 7-year general and seasonal averages at the measurement stations were given in Table 3.

The reason why the highest value according to the 7-year averages was at the 8th station was that the wastewater of Nilüfer and Bursa Organized Industrial Zones was discharged to the Western Wastewater Treatment Plant before this station. The 1st Station, where the minimum concentration was, located in the upstream part of the Nilüfer Stream. The fact that the maximum values were found at the 7th and 8th Stations as a result of the seasonal evaluations shown that these organized industrial zones contribute to the pollution.

When all the pre- and post-pandemic values were analyzed by years and stations, the highest values were measured as 0.0457 mg/L and 0.0675 mg/L at the 10th and 8th Stations in 2020 and 2021, respectively. The minimum value was found to be 0.0127 mg/L at the 9th Station in 2018. The variation graph of the concentrations by years and stations was shown in Figure 2.

EPA and WHO recommended permissible limits for arsenic in drinking water were 0.05 mg/L and 0.01 mg/L, respectively [17,18]. When all stations were examined for 7 years, it was determined that all the values were above the WHO standard, and the annual average of the 8th Station in 2021 was above the EPA standard and all other values were below EPA Standards. Potable water standard value of Turkey (TSE266) was the same as WHO, and none of the measurement stations were in compliance with the Turkish Drinking Water standard [21].

The arsenic limit value allowed in irrigation water was 0.10 mg/L according to the WHO's irrigation water usage guide [22]. Turkish irrigation water standard value was 0.05 mg/L [23]. According to these values, Nilüfer Stream was found to be suitable for irrigation water in terms of As parameter. Only in 2021, the general average of the 8th Station was higher than the Turkish Irrigation Standard.

3.2 Evaluation of Human Health Risk

When the water quality of Nilüfer Stream was evaluated in terms of human consumption risk, it was determined that the hazard quotient (HQ) values were greater than 1 at all stations before and after the pandemic, and therefore there was a chronic and carcinogenic risk. Before the pandemic, the order of magnitude in the stations in terms of HQ values was 10>7>2>8>9>4>3>6>1. The order of HQ magnitude in the stations of post-pandemic was 8>10>7>9>2>6>4>1>3. In general, post-pandemic HQ values and risk had generally increased. A decrease was observed only at the 3rd Station. According to the values before and after the pandemic, it was observed that the 8th and 10th stations had a higher risk, and the 3rd and 1st stations had a lower risk than the other stations.

Table 3. General and seasonal means of 7 Years in the measurement stations

Seasons	Stations									
	1	2	3	4	5	6	7	8	9	10
7 Years mean	0.0182	0.0217	0.0195	0.0202	Not Enough Value	0.0206	0.0239	0.0256	0.0203	0.0213
7 Years Spring mean	0.0187	0.0201	0.0203	0.0209		0.0203	0.0221	0.0196	0.0197	0.0212
7 Years Summer mean	0.0192	0.0229	0.0213	0.0216		0.0210	0.0231	0.0210	0.0203	0.0190
7 Years Autumn mean	0.0181	0.0227	0.0204	0.0204		0.0214	0.0234	0.0309	0.0223	0.0234
7 Years Winter mean	0.0170	0.0214	0.0190	0.0193		0.0201	0.0337	0.0327	0.0190	0.0192

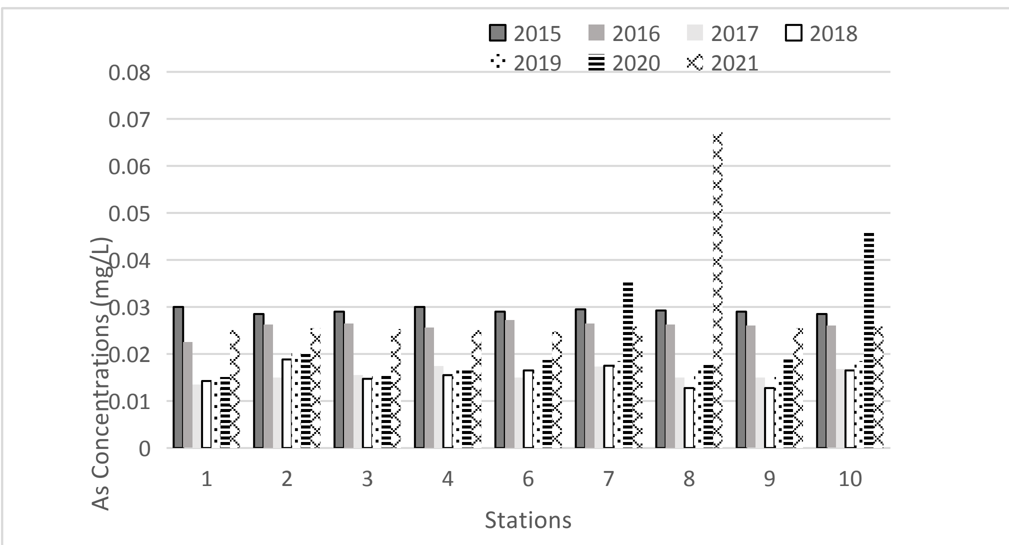

Figure 2. The variation graph of the concentrations by years and stations

The HQ order of the measuring stations was found as 8>7>2>10>6>9>4>3>1 according to the total 7-year averages before and after the pandemic. According to the post-pandemic and general average values, it could be said that the 8th Station had the highest risk, and the 3rd and 1st Stations had low risk.

When all HQ values were examined, it was seen that the highest value was 3.316 at Station 8 after the pandemic, and the lowest HQ value was at Station 1 before the pandemic with 1.615.

When the CR (carcinogenic risk) numbers were examined, it was hazardous for 7 years before and after the pandemic. After the pandemic, the values increased a little more. It was observed that the highest CR value was 14924×10^{-6} at the 8th Station after the pandemic, and the lowest was 743×10^{-6} at the 1st Station before the pandemic.

The 10th and 8th stations, where the concentrations were high, were located after the junction of the side streams and the discharge of Nilüfer and Bursa OIZs. Therefore, the pollution was more in there. The 1st Station was the upstream of the Stream, and the pollution sources were mixed later. Therefore, the health risk was found to be lower than the other stations. ADD, HQ and CR values calculated according to the general averages of 7 years before and after the pandemic were given in Table 4.

4. Conclusions

As a result of all the evaluations, it had been seen that Nilüfer Stream was not suitable for potable water quality according to National and International standards in terms of As parameter, but it was suitable for irrigation water quality.

After the pandemic, a decrease in pollution occurred, which was estimated to be due to the decrease in the production of the industrial zones located in the eastern part of the city only. However, pollution increased at all stations in other parts of the Stream. Therefore, the COVID-19 pandemic did not reduce As pollution. It had increased during the pandemic. It had been thought that the reason for this might be that the wastewater treatment plants did not carry out adequate treatment during the pandemic. Since the concentrations were higher than the standard values and a health risk in the upstream part where the point pollution sources were the least, it was shown that the pollution was not only caused by industry. It was also caused by the natural soil structure of the basin, mining activities and pesticide use. However, the increase in the concentration towards the downstream shown that the industry increased the pollution.

It was determined that the hazard quotient (HQ) values were greater than 1 at all stations before and after the pan-

Table 4. ADD, HQ and CR values calculated according to pre- and post-pandemic and 7-years general averages

Station	Pre-Pandemic			Post-Pandemic			Means of 7 years		
	ADD	HQ	CR	ADD	HQ	CR	ADD	HQ	CR
1	0.49537	1.6512	743×10^{-6}	0.535714	1.785714	804×10^{-6}	0.506667	1.688889	760×10^{-6}
2	0.57197	1.9066	858×10^{-6}	0.605556	2.018519	908×10^{-6}	0.603395	2.011317	905×10^{-6}
3	0.539352	1.7978	809×10^{-6}	0.524306	1.747685	786×10^{-6}	0.542088	1.806958	813×10^{-6}
4	0.540509	1.8017	811×10^{-6}	0.559524	1.865079	839×10^{-6}	0.559722	1.865741	840×10^{-6}
5	Not Enough Measurements								
6	0.534392	1.7813	802×10^{-6}	0.572222	1.907407	858×10^{-6}	0.571429	1.904762	857×10^{-6}
7	0.576389	1.9213	865×10^{-6}	0.883838	2.946128	13258×10^{-6}	0.663105	2.210351	995×10^{-6}
8	0.554167	1.8472	831×10^{-6}	0.994949	3.316498	14924×10^{-6}	0.710573	2.368578	1066×10^{-6}
9	0.543056	1.8102	815×10^{-6}	0.618056	2.060185	92710×10^{-6}	0.564484	1.881614	847×10^{-6}
10	0.590278	1.9676	885×10^{-6}	0.993056	3.310185	1489×10^{-6}	0.590278	1.967593	885×10^{-6}

demic, and therefore there was a chronic and carcinogenic risk. Post-pandemic HQ values and risk had generally increased.

As a result, in order to prevent As pollution, it must to examine in detail the industrial (point) and agricultural (diffuse) pollutant sources originating from pesticide usage and to take protective measures. It must carry out the necessary inspections especially regarding the mining activities in the basin.

Conflict of Interest

There is no conflict of interest.

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